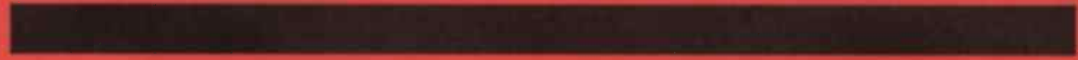


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# Radiocarbon

An International Journal of Cosmogenic Isotope Research



Abstracts of the 7th International Conference  
on Accelerator Mass Spectrometry  
Tucson, Arizona, 20-24 May 1996

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*Editor:* AUSTIN LONG

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*List of laboratories.* Our comprehensive list of laboratories is available upon request. We are expanding the list to include additional laboratories and scientific agencies with whom we have established contacts. The editors welcome information on these or other scientific organizations. We ask all laboratory directors to provide their laboratory code designation, as well as current telephone and fax numbers, and e-mail addresses. Changes in names or addresses, additions or deletions should be reported to the Managing Editor. Conventional and AMS laboratories are now arranged in alphabetical order by country and we include laboratories listed by code designation.

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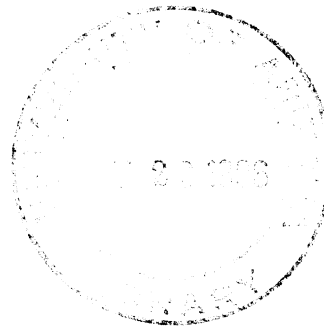
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## INTRODUCTION

This issue of *RADIOCARBON* contains two sets of abstracts. The first set features abstracts from the 7th International Conference on Accelerator Mass Spectrometry (Tucson, 20–24 May 1996) and two associated workshops. AMS-7 continues the tradition of AMS conferences, focusing on the development of the method as well as new sample techniques and important applications of AMS. We also include abstracts from two important workshops highlighting these applications: the workshop on Applications of AMS to Global Climate Change (La Jolla, California, 18 May 1996) and Geological Applications of AMS (Tucson, 25 May 1996). The full papers from the AMS-7 conference will be published in *Nuclear Instruments and Methods in Physics Research*.

The second set of abstracts is from the Workshop on Secular Variations in Production Rates of Cosmogenic Nuclides on the Earth. This conference, held in Santa Fe, New Mexico from 2–5 February 1996, considered the technical aspects and interpretations of one growing application of AMS—cosmogenic radionuclides in geomorphology. An excellent conference summary and overview begins this section (Gosse *et al.* this volume, p. 135), together with an extensive bibliography of publications featuring the *in-situ* production rate method (p. 142).

Both conferences emphasize the diversity of AMS research and its applications. At *RADIOCARBON*, we encourage the publication of new work in the diverse field of cosmic-ray-produced isotopes other than  $^{14}\text{C}$ . By highlighting these two important conferences, we also hope to encourage submissions from these fields to our journal.

*A. J. T. Jull*

*RADIOCARBON Consulting Editor*

*31 March 1996*

## **ACKNOWLEDGMENTS**

The AMS-7 Conference and associated workshops were made possible in part by the support of:

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# Radiocarbon

1996

## ABSTRACTS OF THE 7TH INTERNATIONAL AMS CONFERENCE

### BRAZILIAN ACCELERATOR MASS SPECTROMETRY PROGRAM

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The AMS program at the S. Paulo 9MV Tandem accelerator has been initiated with major emphasis on projects based on the determination of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$  concentrations. A first report on this program was presented at the AMS Conference in Canberra (Tenreiro *et al.* 1994). Since then, progress has been achieved on the improvement of the voltage stability of the 8UD-9MV terminal and the ionic optics, controlled only by electrostatic elements, except for the analyzing magnet. An automatic system for changing beams of different isotopes has been developed, allowing the rejection of the stable beams before they reach the detector placed at the beam direction. The group from the Fluminense Federal University (UFF), in Niteroi, has helped in the design and construction of a new Bragg gas chamber detector, coupled with an E X DE telescope and a time-of-flight system. A new scattering chamber has also been designed at UFF, and it will be fully dedicated to the AMS program. Beams of  $^{26}\text{Al}$  were produced via the  $^{26}\text{Mg}(\text{p},\text{n})^{26}\text{Al}$  reaction;  $^{36}\text{Cl}$  beams were produced from samples prepared from sea water from different places in South America and placed at a Cs-sputtering ion source. Both beams were successfully accelerated and detected. Recently, another group, from the Londrina State University, has joined the Brazilian AMS project. The main research programs related with the AMS technique, in Brazil, are based on Geological and Oceanographical studies.

### REFERENCE

Tenreiro, C. *et al.* 1994 *Nuclear Instruments and Methods in Physics Research B*92.

### AMS SAMPLE HANDLING IN GRONINGEN

*A. T. AERTS-BIJMA, H. MEIJER and J. VAN DER PLICHT*

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As is the case with the Groningen conventional laboratory, the AMS laboratory handles *ca.* 1000 samples annually such as water (DIC/DOC from groundwater and the ocean), marine carbonates, atmospheric  $\text{CO}_2$ , organic deposits such as peat, soils and macrofossils, and the complete spectrum of archaeological datable materials.

The organic samples are combusted in a CN-analyzer, consisting of a flash combustion/purification unit and on-line mass spectrometer. The  $\text{CO}_2$  is trapped cryogenically and transferred to the graphitization setup. We employ one 10-fold and one 15-fold graphitization systems (ranging from 3 to 8

ml), using reduction with hydrogen and Fe as catalyst. With this setup we can handle samples from 150 mg C and up. Finally, the graphite produced is pressed in a target holder, and stored in nitrogen until the measurement.

We have studied various fractionation effects occurring during the graphite production, possible methane production and isotope fractionation (13d, 14d) as a function of amount of CO<sub>2</sub> left. We will also report on various contamination studies.

## PERFORMANCE OF <sup>14</sup>C DATING WITH AMS AT UTRECHT

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Since 1985, more than 5000 samples have been <sup>14</sup>C dated with the Utrecht AMS facility. In this paper the present status of preparation, measurement and data analysis is discussed. A routine procedure is followed for samples of at least 0.5 mg carbon. The <sup>14</sup>C/<sup>12</sup>C and the <sup>13</sup>C/<sup>12</sup>C ratio are measured and normalized on those of the NOX standard. The AMS-measured δ<sup>13</sup>C value is used to monitor the measurement and must agree within 1–2 σ with that from conventional mass-spectrometry. Contamination in preparation and measurement is corrected for on the basis of the average value observed for the blanks (0.18–0.06 pMC). In routine <sup>14</sup>C measurements on (sub)recent materials, the overall precision is *ca.* 0.4%. Measurements on samples containing <200 μg carbon reveal mass dependence in two respects: 1) the <sup>14</sup>C/<sup>12</sup>C of a blank increases with decreasing amount of carbon, and 2) fractionation occurs in the measured <sup>13</sup>C/<sup>12</sup>C and <sup>14</sup>C/<sup>12</sup>C ratios with defects of 2% and 5%, respectively, for samples of 20 μg carbon. This mass-dependent fractionation is ascribed partly to the sample preparation, partly to the AMS measurement. For this reason, “small” samples are compared with a set of standards and blanks with a similar mass range to correct for the mass-effects. In this way, we find uncertainties up to *ca.* 3% for samples of 20 μg carbon.

## AMS PROGRAM AT THE TANDAR ACCELERATOR

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In this work we present the AMS program that is under development using the 20 UD electrostatic accelerator of the TANDAR laboratory at Buenos Aires. The preliminary measurements were done by tuning <sup>14</sup>C beams. The detection system used for this test was a ΔE–E<sub>R</sub> telescope placed at 0°. For the detection of heavier isotopes some developments are being currently implemented in the Quadrupole Dipole Dipole (QDD) magnetic spectrometer at the TANDAR laboratory. A mylar window placed at the entrance of the quadrupole piece defines a volume filled with N<sub>2</sub> inside the spectrometer, where different isobars are expected to follow different trajectories, *i.e.*, reaching the multiwire detector of the spectrometer at different positions. Furthermore, work is also in progress to construct a time-of-flight facility with two microchannel plates prior to the entrance to the QDD spectrometer to distinguish ions with different mass numbers.



## **NEW APPROACH TO NEGATIVE IONS: LASER PHOTO DETACHMENT AND RESONANT IONIZATION SPECTROSCOPY**

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Studies of the structure and dynamics of negative ions are of fundamental interest for atomic physics, but have also practical implications, such as providing the basis for ultra-sensitive accelerator mass spectroscopy (AMS). Negative ions exhibit structural properties markedly different from neutral atoms because of the extra electron being bound in a short-range potential and the important role played by the electron correlations. During the last year, significant experimental progress has been achieved by means of a newly developed technique based on resonance ionization spectroscopy, which will be reported with examples of its application.

The new method is based on laser photo detachment of the negative ion, followed by state-selective resonance ionization of the generated neutral atom by means of photo excitation into a Rydberg level, with subsequent state-selective field ionization. The sensitivity and selectivity of this technique make it possible to study even very weak photo detachment channels and to rely on s-electron thresholds in the measurements of electron affinities for nearly all elements. The technique has been used to study the structural properties (binding energies, fine-structure splittings, existence of meta-stable state or resonances) of the weakly bound alkaline-earth ions, which have attracted so much interest in recent years. The experimental binding energies and fine-structure splittings are improved by nearly two orders of magnitude compared to recent results yielding data, which has made it possible to perform critical tests of theoretical predictions. It has also been possible to unambiguously clarify the nature and state composition of negative beams, to disprove previous claims to observation of a meta-stable component in  $\text{Ca}^-$ , and to discover the long-lived unpredicted metastable  $\text{Ba}^-$  ion.

## **IMPURITY DETECTION IN THE SEMICONDUCTOR INDUSTRY: CHALLENGES FOR AMS**

*MARK ANTHONY*

Texas Instruments, Dallas, Texas, USA

The semiconductor industry is growing at a rapid rate, with worldwide sales of >\$100 billion in 1995. Although compound semiconductors satisfy some niche roles in the industry, Si is still by far the dominant material in terms of market share. Many technical challenges must be addressed to successfully scale Si critical dimensions to the 250 nm regime in mainstream manufacturing. In many cases, successfully meeting these challenges requires detailed knowledge of the amount and location of impurity elements in Si structures at low concentrations, both laterally and in depth. This presentation will attempt to identify some of the technical needs for impurity detection in the semiconductor industry, and to compare the strengths and weaknesses of AMS with other trace element methods under consideration.

**DETECTION OF  $^{41}\text{Ca}$  WITH  $^7\text{Li}^{37}\text{Cl}$  MOLECULAR PILOT BEAM**

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The dating of foraminifera (Nagashima, Baba *et al.*, in press), which is one of our current interests, is extensively studied by  $^{14}\text{C}$  AMS technique with  $^7\text{Li}_2$  pilot beam (Nagashima *et al.* 1994). As the next item on the dating program, dating of foraminifera with  $^{41}\text{Ca}$  has been tried. A long half-life of  $^{41}\text{Ca}$  enables us to investigate very old foraminifera (several hundred thousand years old). Expanding the idea of the molecular pilot beam method used in  $^{14}\text{C}$ ,  $^{26}\text{Al}$ , and  $^{36}\text{Cl}$  measurements (Nagashima *et al.* 1994), a  $^7\text{Li}^{37}\text{Cl}$  molecular pilot beam has been introduced for stabilizing the terminal potential of  $^{12}\text{UD}$  pelletron machine.

$^{41}\text{Ca}$  standard was prepared with the  $^{40}\text{Ca}$  neutron capture process in a nuclear reactor.  $^{41}\text{Ca}$  to  $^{40}\text{Ca}$  ratio was controlled to be 1.0510–10. Under the chemical process, the form of  $\text{CaCO}_3$  changes to  $\text{CaF}_2$  in order to diminish oxygen. The  $\text{CaF}_2$  sample is mixed with  $\text{TiH}_2$ , and  $\text{LiCl}$  and the mixture is pressed into the hole of an ion source sample holder. A  $\text{CaH}_3^-$  negative beam was created in the ion source as well as the  $^7\text{Li}^{37}\text{Cl}$  molecular pilot beam. The strong  $^{41}\text{K}$  background can be greatly decreased because there is no  $\text{KH}_3^-$  formation in the ion source.  $\sim 1\text{mA}$   $\text{CaH}_3^-$  current can be extracted and  $^{41}\text{CaH}_3^-$  and  $^7\text{Li}^{37}\text{Cl}^-$  are transported into the  $^{12}\text{UD}$  through a 90-degree inflection magnet. After passing through the tandem, both  $^{41}\text{Ca}^{10+}$  and  $^{37}\text{Cl}^{9+}$  are analyzed by an analyzing magnet. Here, the  $^{37}\text{Cl}^{9+}$  beam can produce enough current on a pair of off-axis slits that are set at the image plane of the magnet and the current is used for terminal potential control. On the other hand,  $^{41}\text{Ca}^{10+}$  is guided into an AMS beam line and it is finally detected by a gas DE-SSD E counter. Overall beam transport efficiency is *ca.* 2.5%. We will report the method of  $^{41}\text{Ca}$  detection with the  $^7\text{Li}^{37}\text{Cl}$  molecular pilot beam in detail as well as some preliminary results of  $^{41}\text{Ca}$  measurements.

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Nagashima, Y., Baba, T. *et al.*, in press, *Proceedings of 7th International Conference on Heavy Ion Accelerator Technology*. Canberra, September 1995.

**USE OF CHLORINE-36 AS TRACER FOR THE EVOLUTION OF WATERS IN GEOTHERMAL AND TECTONIC ACTIVE AREAS IN WESTERN TURKEY**

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*and*

*H.-A. SYNAL*

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Within the joint, interdisciplinary project "Marmara" of ETH Zürich and the Istanbul Technical University (ITÜ), six areas along the North Anatolian Fault Zone (Schindler *et al.* 1993; Straub and Kahle 1994) are studied. In particular, effects of active tectonics as evidenced by geology, geodesy and seismology on groundwater circulation and on heat flow pattern are investigated. For groundwaters of such tectonic active regions the problem of residence time determination arises, as the usu-

ally applied isotope techniques are often not appropriate because of 1) the high amount of water-rock interaction; 2) the underground production of nuclides in crystalline, fissured rocks in geothermal conditions; 3) the occurrence of highly saline waters at boiling temperature affected by water-rock interaction processes; and 4) the high fluxes of CO<sub>2</sub>. Therefore, the application of the <sup>36</sup>Cl method looks very promising. We discuss the results of <sup>36</sup>Cl determinations of groundwaters from six areas along the North Anatolian Fault zone: (from East to West) Kuzuluk/Adapazari, Yalova/Gemlik, Bursa, Gönen/Yenice, Bergama and Canakkale (Tuzla/Kestanbol).

Main subsurface effects that affect <sup>36</sup>Cl concentration of groundwaters in tectonic active areas are: 1) underground production; 2) leaching of rock salt (including fluid inclusions) due to water-rock interaction processes; 3) dissolution of limestones, especially in areas of high CO<sub>2</sub> activity related to thermometamorphic processes; and 4) mixing processes of fluids with different origin/evolution with different chloride concentrations and <sup>36</sup>Cl/Cl ratios. Considering for each area the sources of <sup>36</sup>Cl and the water-rock interactions with respect to the chloride cycle, additional information on the ongoing processes of groundwater circulation and evolution as well as the ranges of residence times was gained. Dating with <sup>36</sup>Cl is possible only if the local input ratio and exact evolution (from the rock environment along flow path including the geological evolution of the area) is known. Additional information, such as independent hydrodynamic and coupled geothermal modeling as well as the results of other environmental isotopes, gives additional support to the interpretation described above.

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#### APPLICATION OF THE <sup>36</sup>Cl METHOD FOR THE INVESTIGATION OF THE MULTI-LAYER AQUIFER SYSTEMS OF THE GREAT HUNGARIAN PLAIN (HUNGARY)

W. BALDERER,<sup>1</sup> H-A. SYNAL,<sup>2</sup> J. DEÁK<sup>3</sup> and W. STICHLER<sup>4</sup>

The Great Hungarian Plain (GHP), with an areal extension of >50,000 km<sup>2</sup>, is a sedimentary basin in Central/Eastern Hungary. This deep depression basin was formed mainly in the late Tertiary and Quaternary. The continuous subsidence of the basin led to the accumulation of sedimentary layers of several thousands of meters of thickness. This area is well defined with respect to hydrogeology by its large density of wells, and has been carefully studied with respect to hydrochemistry and environmental isotope techniques, including noble gases (Deák *et al.* 1987; Stute 1989). It was therefore selected as appropriate for further investigations by the <sup>36</sup>Cl method. Groundwater samples were collected along four different flow paths starting from the recharge areas at the southern, eastern and northwestern part and leading to the central part of the main discharge area of the basin. The groundwater samples originate from boreholes with defined depth intervals (as the screened part of the boreholes) within the sedimentary layers of the Quaternary aquifer (used mainly for drinking water supply of the towns on the plain) and the underlying thermal water aquifer of the Pannonian (late tertiary sediments of the Upper Pliocene). From the first evaluation of the data of the groundwaters sampled in August 1995 the following observations result: along the flow path an increase in chloride concentration together with generally decreasing <sup>36</sup>Cl concentrations (<sup>36</sup>Cl atoms/l of water)

can be recognized. Generally, groundwaters originating from boreholes at greater depths at the same location show lower  $^{36}\text{Cl}/\text{Cl}$  ratios.

For some samples of boreholes from the same location an inverse trend is observed. Such situations are possibly linked to mixing phenomena due to the continuous exploitation of the groundwater within the quaternary aquifer as a drinking water supply. The residence times deduced from the  $^{36}\text{Cl}$  results (by taking into account the chemical evolution of the waters and also the amount of underground production) will be discussed for the different flow paths and compared to the results of other, previously applied isotope methods.

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## $^{14}\text{C}$ /AMS SINGLE-RING ANALYSIS OF A LATE PLEISTOCENE TASMANIAN CELERY-TOP PINE

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Ancient conifer logs are exposed in the bed and banks of the Stanley River in western Tasmania. Many of them have been sampled for dendrochronology and  $^{14}\text{C}$  studies. In addition, several major excavations have been carried out in floodplain sediments up to 100 meters from the present river channel. Sections have been collected from over 350 well-preserved subfossil logs. They range in age from >48 ka to the present, with good coverage for the periods 9 ka to 3.3 ka and from 2.5 ka to the present. Two species are represented in the collection, Huon pine (*Lagarostrobos franklinii*) and celery-top pine (*Phyllocladus aspleniifolius*), with the former more abundant.  $^{14}\text{C}$  measurements have previously been made on consecutive 10-ring samples from several logs, using classical liquid scintillation methods (Barbetti *et al.* 1992, 1995). We recently measured with a precision of 0.5 to 1% the  $^{14}\text{C}$  concentrations of 77 consecutive, single-ring samples from a celery-top pine log, catalogue SRT-157, dating from ca. 12,700  $^{14}\text{C}$  yr BP. The rings were separated by careful shaving, then milled and pretreated to isolate alpha-cellulose before combustion to  $\text{CO}_2$  and graphitization. These data provide a short but precise high-resolution annual record of atmospheric  $^{14}\text{C}$  variations when plotted against ring numbers, and will be used to estimate an upper limit for the amplitude of the 11-yr solar cycle.

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## STUDIES ON THE UPTAKE OF $^{26}\text{Al}$ INTO A CULTURED BLOOD-BRAIN BARRIER

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Aluminium has been implicated in the aetiology of Alzheimer's Disease (a form of dementia). It has been suggested that silicon may play a role in suppressing the uptake of aluminium into the brain by the formation of stable aluminosilicates. These complexes are also present in the cores of certain histopathological lesions (Neuritic Plaques) in patients with Alzheimer's Disease (AD). However, it is not known whether the distribution of these elements is an artifact from the measurement procedure or a genuine co-localization. Recently, it has been shown, using accelerator mass spectrometry (Edwardson *et al.* 1993; Barker *et al.* 1992; Day *et al.* 1991; 1994) that silicon in the diet can reduce the gastrointestinal uptake of aluminium in humans and that most aluminium is bound to the iron-transporting protein transferrin in blood plasma. There is often a negative correlation between aluminium and silicon in potable water supplies, and the reported geographical association between AD and aluminium from tapwater may be a consequence of dissolved silicon promoting the formation of insoluble aluminosilicates. The synergic behavior between silicon and aluminium in biological systems is poorly understood and there is an urgent need to study both the uptake and the Al/Si interaction at the brain endothelium (Blood-Brain Barrier).

Fully grown cells were incubated with  $^{26}\text{Al}$  and stable aluminium carrier in the medium for up to 4 hr in the presence and absence of transferrin, and in the presence of an antibody which blocks the interaction of the transferrin-aluminium complex to its receptor. To investigate the role of aluminium speciation, aluminium citrate and silicic acid were also used.

At the end of the time period, the medium was removed, cells washed and samples prepared for counting. The full results of these studies will be presented at the meeting.

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**A NEGATIVE SURFACE IONIZATION SOURCE FOR AMS***M. BAUER, D. DEBOFFLE, R. MEUNIER, G. RAISBECK and F. YIOU*

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For isotopes with large electronegativities ( $^{129}\text{I}$ ,  $^{36}\text{Cl}$ ) a negative ion source based on surface ionization is potentially very attractive, both from the point of view of efficiency, and/or selectivity from interfering isobars. We have constructed a prototype source based on this principle, using a heated surface of  $\text{LaB}_6$  or  $\text{BaO}$ . We have carried out preliminary tests of the source on the isotope separator SIDONIE using samples prepared from stable iodine ( $\text{AgI}$ ).

The results demonstrate the feasibility of using small samples ( $<60 \mu\text{g}$  iodine) and efficiencies of  $>1\%$ . This opens up the possibility of measuring  $^{129}\text{I}/^{127}\text{I}$  ratios directly from 1 liter ocean water samples, without addition of carrier.

 **$^{10}\text{Be}$  AND DUST IN POLAR ICE***ST. BAUMGARTNER,<sup>1</sup> J. BEER,<sup>1</sup> G. WAGNER,<sup>1</sup> P. KUBIK,<sup>2</sup> M. SUTER,<sup>3</sup> G. M. RAISBECK<sup>4</sup> and F. YIOU<sup>4</sup>*

The dust content of today's precipitation in the polar regions of the Earth is so small that the contribution of recycled, dust-bound  $^{10}\text{Be}$  to the total  $^{10}\text{Be}$  signal in polar snow and ice is generally regarded to be negligible. During the last ice age, however, dust fluxes were larger than today by at least one order of magnitude. One is therefore forced to examine the relevance of dust as a source of  $^{10}\text{Be}$  in polar ice cores. New experimental data from the Summit GRIP ice core suggest that the ratio of dust adsorbed  $^{10}\text{Be}$  to the total  $^{10}\text{Be}$  content is much larger than expected, since the  $^{10}\text{Be}$  concentration of the dust is very high. The dust contribution to the  $^{10}\text{Be}$  signal, therefore, cannot be neglected. Its knowledge is necessary for a thorough interpretation of the whole  $^{10}\text{Be}$  record.

These unexpected findings lead to some fundamental questions about possible interferences between the two sources of  $^{10}\text{Be}$  in precipitation, *i.e.*, dust with recycled  $^{10}\text{Be}$  and aerosols with recently produced  $^{10}\text{Be}$ . The question has to be addressed, under which conditions the two sources for  $^{10}\text{Be}$  can be separated at all. A status report on actual problems and possible solutions will be given.

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In this paper we will present a summary of the activities at the NSF-Arizona AMS laboratory over the past year. In addition to a description of some of the measurements, the summary will include details of: 1) latest results concerning precision and accuracy of radiocarbon measurements; 2) mea-

surements of, and corrections for,  $\delta^{13}\text{C}$  values; 3) corrections to  $^{14}\text{C}$  measurements for contamination introduced in sample preparation; 4) dependence of  $^{14}\text{C}$  ages on sample size and target composition; 5) properties of  $^{10}\text{Be}$  measurements; 6) a recent modification to the accelerator; and 7) planned future additions to the instrument. The particular modification to be discussed is the installation and operating characteristics of an insulator which supports the high-voltage terminal. This insulator allows us to decrease the stresses on the delrin rods which are used to maintain the accelerator tubes under compression.

### HIGH FREQUENCY RADIOCARBON VARIATIONS IN PACIFIC CORALS

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Modern corals from the western Pacific Ocean reveal substantial seasonal cycles in  $\Delta^{14}\text{C}$  when sampled at a monthly frequency. GCM model results suggest that a significant portion of these seasonal cycles may be advected from the equatorial Pacific region, as opposed to being of local origin.

Coupled U/Th and  $^{14}\text{C}$  measurements on a long-lived (700 yr) Younger Dryas (YD) age coral reveal periodic inter-annual to decadal scale variability in ocean surface water  $\Delta^{14}\text{C}$ , with variations of 10–40‰ amplitudes. These periodic variations are superimposed on a long period 150‰ drop in  $\Delta^{14}\text{C}$  which occurred during the YD. Carbon box model results suggest that such changes cannot plausibly be produced by changes in the Earth's magnetic field intensity, but that it is possible to produce such variations solely by modulating the thermohaline circulation flux associated with NADW.

### $^{36}\text{Cl}$ VARIATIONS IN GREENLAND ICE SINCE 1425 AD

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$^{36}\text{Cl}$  was measured in 470 samples of a 300-m-long ice core from Dye 3, Greenland, which has been analyzed previously for  $^{10}\text{Be}$ . The  $^{36}\text{Cl}$  record has an annual resolution and reveals interesting features. The solar modulation is reflected in the Schwabe sunspot cycle as well as in the 90-yr Gleissberg cycle. The Maunder and the Spörer minima are also present. In the 1950s the  $^{36}\text{Cl}$  concentration was increased dramatically by nuclear bomb tests.

A detailed comparison between  $^{36}\text{Cl}$  and  $^{10}\text{Be}$  measured on the same pre-bomb samples shows a relatively high similarity of the basic features of both records. However, on the average  $^{36}\text{Cl}$  lags behind  $^{10}\text{Be}$  by about one year. This points to a longer atmospheric residence time of  $^{36}\text{Cl}$  compared to  $^{10}\text{Be}$  and provides some hints to atmospheric transport processes.

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### WEAKLY BOUND NEGATIVE IONS STUDIED BY LASER EXCITATION AND AMS

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Properties of weakly bound elemental and molecular negative ions (electron affinity and photodetachment cross section) are measured by combination of laser excitation and AMS techniques. The photodetachment cross section is determined by measuring the depletion of negative ions through

laser light. The electron affinity is obtained by varying the photon energy and studying the excitation function near a threshold corresponding to a state in the neutral atom. The negative ion beam formed in a Cs<sup>+</sup> sputter source is alternately transmitted after colinear interaction with a 25 Hz dye laser pulse or with no laser interaction. Transmitted ions are analyzed by AMS and counted. For elements with very small affinities, the yield of negative ions is usually weak due either to low probability of formation or to other effects (*e.g.*, decay by black-body radiation). When, in addition, the ion mass is large, the ion beam contains usually high-intensity backgrounds of negative molecular ions of equal mass number. The use of AMS to tag the ion species interacting with the laser pulse is then essential. The energy of individual laser pulses corresponding to every detected ion is measured, allowing for a direct measurement of the absolute photodetachment cross section. An alternative technique designed to make measurements close to threshold (small cross sections) more sensitive, is based on the detection of the neutral reaction products rather than the depleted negative ion beam. Negative ions interact with the laser pulse in the tandem terminal and neutral products are separated from the negative ion beam and identified by AMS after a subsequent stripping along the high-energy accelerating tube. We measured absolute photodetachment cross sections of Sr<sup>-</sup> ions interacting with photons at energies near the threshold corresponding to the first excited state of the neutral Sr atom. By fitting a theoretical law to the measured excitation function an electron affinity of  $48 \pm 6$  MeV has been determined (Berkovits *et al.* 1995). Negative ions of lanthanide and actinide elements are under investigation. The experimentally measured electron affinities and the absolute cross sections are a guide to theoretical atomic calculations since they constitute a sensitive test of the accuracy of the wave functions used. This is especially true for the group of the lanthanide and actinide elements, where very little experimental information exists.

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#### RECENT EXAMPLES OF <sup>14</sup>CO-AMS APPLICATIONS IN THE STRATOSPHERE AND TROPOSPHERE

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Good progress has been made in the application of AMS in atmospheric chemistry. After the extensive, more traditional measurements of atmospheric carbon dioxide using conventional radiocarbon dating, methane <sup>14</sup>C levels have been established using AMS, and presently this technique allows us to obtain <sup>14</sup>CO observations. Using air samples of some several hundred liters of air, it has been established that the lowest levels occur, as expected, in the troposphere, where summertime minima of 5 molecules <sup>14</sup>CO per cm<sup>3</sup> of air were detected. In the lowermost high latitude stratosphere the highest recorded level so far is 200 molecules <sup>14</sup>CO per cm<sup>3</sup> of air (in descending polar vortex air at 200 hPa). This enormous gradient offers some exiting applications. In particular it will give an unprecedented test for our quantitative estimates of the intensity of troposphere-stratosphere exchange, which traditionally has been repeatedly discussed in relation with the flux of ozone from the stratosphere into the troposphere. In fact the good old <sup>14</sup>CO<sub>2</sub> (carbon-14 dioxide) from the atmospheric bomb tests has been one of the few benchmarks for estimating stratospheric residence times so far.



However, a most exciting application is the assessment of tropospheric OH, which is based on the fact that the dominant sink of  $^{14}\text{CO}$  is OH, via  $^{14}\text{CO} + \text{OH} (^{14}\text{CO}_2 + \text{H})$ . This OH radical is extremely short lived and it defies large-scale direct measurement, yet it is fundamental to the photochemical cleaning process of the atmosphere. The first results we have obtained showed that probably the established OH values had been too low by *ca.* 20%, and indeed, just recently the independent estimates for OH have been “re-adjusted”. This meant, for instance, that the lifetime of the greenhouse gas methane in the atmosphere has been scaled down from *ca.* 9 to 8 yr. Although this fact has almost gone unnoticed, we are looking forward to a most rewarding application of  $^{14}\text{CO}$  which will help to better constrain 1) stratosphere-troposphere exchange, 2) meridional transport, and 3) tropospheric OH levels. We will present and discuss the latest results in the use of  $^{14}\text{CO}$ .

### **CORRECTIONS FOR CONTAMINATION BACKGROUND IN AMS $^{14}\text{C}$ MEASUREMENTS**

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Measurements of  $^{14}\text{C}/^{13}\text{C}$  ratios of a standard material (HOxI) and  $^{14}\text{C}$  “dead” materials have been made that span the mass range from *ca.* 10  $\mu\text{g}$  to 2 mg. These measurements have allowed the determination of both the amount of the contaminant carbon introduced during sample processing in our laboratory and  $^{14}\text{C}$  content of the contaminant carbon. These data have been used to correct  $^{14}\text{C}/^{13}\text{C}$  ratios obtained for a test sample *ca.* one half-life old for influence of the background contaminant. The  $^{14}\text{C}/^{13}\text{C}$  ratios measured for the test sample span the 10  $\mu\text{g}$  to 2 mg mass range and the corrections have been made using three different formulae. The results obtained from these calculations allow the accuracy of these background correction formulae to be evaluated.

This work was performed under the auspices of the U.S. Department of Energy at the Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

### **A RADIOCARBON CALIBRATION DURING THE LAST DEGLACIATION BASED ON TIMS $^{230}\text{Th}$ AGES OF AMS $^{14}\text{C}$ DATED CORALS FROM VANUATU, NEW HEBRIDES**

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Radiocarbon calibration beyond the range of tree rings is critical not only to improve the accuracy of  $^{14}\text{C}$  ages and correlations, but also because the pattern of secular variation gives important clues about the nature of deglacial climate transitions. Previous dating studies of corals have demonstrated that  $^{230}\text{Th}$  ages give an independent calibration of the  $^{14}\text{C}$  time scale beyond the limits of dendrocalibration. We present a comparison of high-precision thermal ionization mass spectrometry (TIMS) U/Th ages to AMS  $^{14}\text{C}$  ages of corals drilled from an uplifted reef along the coast of Tasmaloum, Espiritu Santo, Vanuatu. Tectonic uplift of the coastline provided a substrate for a relatively continuous sequence of reef growth during periods of rapid post-glacial sea-level rise between 14 and 7 thousand years ago during the transition from full glacial to full interglacial conditions. The Vanuatu coral calibration indicates an overall pattern of decreasing  $^{14}\text{C}/^{12}\text{C}$  throughout the last deglaciation. However, superimposed upon this gradual trend, the Vanuatu calibration shows two prominent  $^{14}\text{C}$  plateaux/reversals that are temporally correlated with two  $^{14}\text{C}$  plateaux identified in terrestrial varved  $^{14}\text{C}$  calibrations. Assuming a constant reservoir age of the surface waters, these features would suggest a rapid decrease in atmospheric  $^{14}\text{C}/^{12}\text{C}$  (from  $\sim 250\text{‰}$  to  $130\text{‰}$ ) between

14,800 ± 100 and 14,150 ± 70 <sup>230</sup>Th yr BP, followed by a rise in  $\Delta^{14}\text{C}$  to ~200‰ by 12,600 ± 60 <sup>230</sup>Th yr BP, followed by a second large drop in  $\Delta^{14}\text{C}$  of ~120‰ between 12,300 and 11,000 yr BP. Local or regional changes in the reservoir age may account for part of the observed  $\Delta^{14}\text{C}$  change. However, the fact that these drops are temporally correlated with independent evidence of NADW “turn on” at 14,700, and 12,500 yr BP and two distinct periods of rapid warming recorded in ice cores and terrestrial paleoclimate records suggests that global changes in ocean circulation were an important contributing factor. The high magnitude and rapidity (>80‰ in <800 yr) of these  $\Delta^{14}\text{C}$  shifts could result from rapid ventilation and outflow of <sup>14</sup>C-depleted water from the Southern Ocean into the South Pacific thermocline following long periods of reduced outflow and decay of oceanic <sup>14</sup>C trapped in the deep waters around Antarctica. These ventilation events may be induced by increases in the flux of NADW and/or shifts in the latitude of the maximum southern-hemisphere westerly wind belt. The results of this study underscore the need for high-resolution studies of  $\Delta^{14}\text{C}$  variation from around the world during these specific time intervals.

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## **RADIOCARBON DATING OF DISSOLVED ORGANIC AND INORGANIC COMPONENTS IN GROUNDWATER**

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This study compares radiocarbon dates and  $\delta^{13}\text{C}$  measurements of dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC) from the same groundwater samples. Water samples were taken from several carbonate aquifers in southern Nevada. Dissolved inorganic fractions were separated by acidifying the water under a vacuum. Dissolved organic fractions were collected with an Amicon 8400 stirred cell ultrafiltration unit. Subfractions of low molecular weight (LMW), middle molecular weight (MMW) and high molecular weight (HMW) were collected using Amicon Diaflo YM1 and YM3 ultrafilters. The LMW fraction contained material with molecular weights below 1000 amu. The molecular weight of the MMW fraction ranged from 1000 to 3000 amu and the HMW fraction contained material with molecular weights >3000 amu. Each of these fractions was concentrated with the ultrafiltration unit, freeze-dried, combusted, and reduced to graphite for AMS analysis.

Results suggest that the HMW fraction is the best DOC component to use for <sup>14</sup>C dating. Dates derived from the HMW fraction are broadly consistent with dates derived from hydraulic models and from chemical mass balance models based on DIC measurements. For younger samples (less than a few 100 years), the <sup>14</sup>C concentration of the DIC fraction is higher than that of the HMW fraction, but for older samples, the opposite relationship is observed. Details of the technique will be discussed, with a comparison of this method to published studies using other methods to isolate DOC in groundwater.

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## INVESTIGATION OF THE QUATERNARY HISTORY OF THE COLORADO RIVER USING COSMOGENIC RADIONUCLIDES

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Cosmic-ray-produced radionuclides in surface samples are being used to investigate the emplacement of colluvial and fluvial deposits in the eastern Grand Canyon, between Lee's Ferry and Unkar Creek. The colluvial surfaces in this reach of the Colorado River are mantled by debris-flow deposits emplaced by tributary washes or on pediments, whereas the fluvial surfaces are comprised of river gravels. Downcutting of the Colorado River punctuated by periods of stasis or aggradation resulted in the formation of multiple terraces in which the lowest levels are the youngest. The terrace levels were identified on the basis on morphostratigraphy. These surfaces are mantled by locally derived rocks, many of which contain chert. Cosmogenic <sup>10</sup>Be and <sup>26</sup>Al are produced by cosmic ray interactions in chert so these samples should help to establish a chronology for aggradation events in this portion of the Grand Canyon. Soil samples were also obtained from most surfaces sampled for cosmogenic radionuclide measurements, providing a means for calibrating and constraining models of carbonate formation.

In the samples analyzed to date we have demonstrated that chert is a suitable mineral for surface exposure dating. The surfaces have exposure ages ranging from <2000 yr to >100 ka and the lowest level surfaces yield the youngest ages. Based on the ages of the youngest colluvial surfaces, which are dated by archaeological methods, and on the overall consistency of the surface exposure ages, we believe that most samples do not contain inherited cosmogenic <sup>10</sup>Be and <sup>26</sup>Al. The older surfaces show evidence of degradation and yield exposure ages less than would be indicated by their stratigraphic level.

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## TRACING THE ENVIRONMENTAL EFFECT OF NUCLEAR-WASTE DISPOSAL TO THE OB RIVER SYSTEM IN CENTRAL SIBERIA, RUSSIA USING ACCELERATOR MASS SPECTROMETRY

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In May and June 1995, a Russian-American research team sampled the flowing Ob River system, including the suspended and bed sediments, for determination of radionuclide concentrations. The research team consisted of seven Russian scientists from the State Hydrologic Institute, St. Petersburg, and four hydrologists from the U.S. Geological Survey (USGS). Water samples were collected using high-volume peristaltic pumps, then were prefiltered through 63-micrometer ( $\mu\text{m}$ ) nylon media and filtered sequentially through 0.45- $\mu\text{m}$  and 10,000-dalton tangential-flow ultrafiltration systems. Samples were collected from five stations along the Ob River system representing channel length of >3,500 km from the headwaters in central Siberia to the Ob Gulf on the Kara Sea. Two

known nuclear-weapons production facilities on this river system are the probable sources of the radionuclides measured.

Chlorine-36 ( $^{36}\text{Cl}$ ) concentrations were determined by accelerator mass spectrometry (AMS) for 500 mL (milliliter) water samples prefiltered through the 63- $\mu\text{m}$  nylon media. Concentrations of  $^{36}\text{Cl}$  were measured at all five stations and ranged from  $4.0 \pm 0.1 \times 10^7$  atoms/L for the sample only influenced by nuclear-weapons testing and not by waste disposal (station 1) to  $2.4 \pm .03 \times 10^9$  atoms/L for the sample nearest a plutonium production facility (station 2). The  $^{36}\text{Cl}$  concentration for the sample from the station most distant from any waste source and nearest to the Ob Gulf (station 5) was  $5.0 \pm 0.2 \times 10^8$  atoms/L.

Plutonium (Pu) isotopic concentrations and strontium-90 ( $^{90}\text{Sr}$ ) concentrations were determined by conventional decay-counting methods. The only Pu isotopes detected in 20 L water samples were  $^{238}\text{Pu}$  at station 1 and  $^{239}\text{Pu}$  at station 2. No detectable concentrations of  $^{90}\text{Sr}$  were measured in 400 mL water samples from any of the stations. Gamma-spectroscopy measurements also showed no detectable concentrations of anthropogenic gamma-emitting radionuclides from the 20 L water samples. These results suggest that AMS offers a method of determining radioactivity concentrations in the environment at greater distances away from a source than do conventional decay-counting methods because of the smaller analytical method detection limit associated with AMS for select radionuclides.

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## **BOMB-PRODUCED CHLORINE-36 FLUX CALCULATED FROM MID-LATITUDE GLACIAL ICE OF NORTH AMERICA**

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In 1991, the U.S. Geological Survey collected a 159.7-m ice core from the Upper Fremont Glacier in the Wind River Range, Wyoming (Naftz *et al.* 1993). In 1994, the ice was processed at the National Ice Core Laboratory in Denver, Colorado, and analyzed for chlorine-36 ( $^{36}\text{Cl}$ ) by accelerator mass spectrometry at Purdue University. A tritium bomb peak identified in the work by Naftz and others was used as a marker to estimate the depth of bomb-produced  $^{36}\text{Cl}$ . Tritium concentrations ranged from 0 tritium units (TU) for older ice to >300 TU at 29 m below the ice surface, a depth that includes ice that was deposited during nuclear-weapons tests through the early 1960s. Maximum  $^{36}\text{Cl}$  production during nuclear-weapons tests was in the late 1950s; therefore, the analyses were performed on ice from a depth of 29.8 to 32 m. Calculated flux for  $^{36}\text{Cl}$  in ice deposited in the late 1950s ranged from  $4.5 \pm 0.1 \times 10^6$  atoms/cm<sup>2</sup> yr for a 0.6-m section of ice centered at a depth of 30.1 m to  $11 \pm 0.2 \times 10^6$  atoms/cm<sup>2</sup> yr for a 0.5-m section of ice centered at a depth of 31.8 m. Ice samples from a depth of 104.7 to 106.3 m were selected to represent pre-weapons tests  $^{36}\text{Cl}$  flux. Calculated flux for  $^{36}\text{Cl}$  in this deeper ice was  $1.7 \pm 0.2 \times 10^5$  atoms/cm<sup>2</sup> yr for a 0.8-m section of ice centered at a depth of 105.1 m and  $7.4 \pm 0.4 \times 10^5$  atoms/cm<sup>2</sup> yr for a 0.8-m section of ice cen-

tered at a depth of 105.9 m. These flux calculations from the Upper Fremont Glacier analyses are the first for bomb-produced  $^{36}\text{Cl}$  in ice from a mid-latitude glacier in North America.

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#### $^{14}\text{C}$ ANALYSIS ON THE SINR MINI-CYCLOTRON AMS

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Since the completion of the first phase of the minicyclotron AMS in 1993, the facility SMCAMS has been steadily operated and greatly improved. To put this facility into practical use, endeavors in three areas have been made. The home-designed multi-sample device for the vertical Cs sputter negative ion source has been assembled on the injector of the minicyclotron and has been working routinely. The "slow" alternate acceleration of  $^{12}\text{C}$ ,  $^{13}\text{C}$  and  $^{14}\text{C}$  has also preceded, and has showed that the read-out of the three kinds of particles is reproducible, which implies machine performance is stable. Five new power supplies for "fast" alternate acceleration have been delivered to our laboratory, but haven't been tested yet. In the meantime, we spent a lot of energy in increasing the  $^{14}\text{C}$  counts to >5 cps from 1–1.5 cps for modern samples made from sugar by finely adjusting the machine, redesigning the accelerating electrodes, improving the vacuum and further suppressing secondary electron and X-ray interference to the micro-channel plate detector. We are feeling confident of further increasing the  $^{14}\text{C}$  counts to well above 10 cps, if some tested techniques (such as a buncher) can be applied and some explored problems can be solved.

Since early this year, the  $^{14}\text{C}$  analysis has proceeded on this facility, and the optimum method of  $^{14}\text{C}$  analysis on the SMCAMS has been explored arising from the different  $^{14}\text{C}$  analysis principle between tandem AMS and minicyclotron AMS. In this paper, improvements of the facility will be summarized, the results of  $^{14}\text{C}$  measurements will be presented and analysis principle will be described. Finally, the potentials of this facility will be highlighted.

**ANIMAL STUDIES OF Al METABOLISM WITH AMS TECHNIQUE**

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Animal tests have been performed to study the correlation of Al metabolism and senile dementia. Four groups of lab rats were fed with normal food, normal food with low and high aluminium contents and high aluminium content plus calcium and magnesium respectively for six to 12 months. At the end of this period, a mapping test was made to record the degree of cognitive degeneration. Half of the rats were sacrificed and Al contents in serum and various organs measured by atomic absorption spectroscopy. The other half were injected with  $^{26}\text{Al}$ , killed after 5, 10, 15, 25 and 35 days and  $^{26}\text{Al}$  contents measured by AMS. The distribution of Al and  $^{26}\text{Al}$  was analyzed and the correlation of the accumulation of  $^{26}\text{Al}$  with the initial Al content and dementia was studied.

**MEASUREMENT OF  $^{81}\text{Kr}$  IN THE ATMOSPHERE**

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We present the first AMS measurement of the  $^{81}\text{Kr}$  concentration in atmospheric krypton. Our result,  $^{81}\text{Kr}/\text{Kr} = (5.3 \pm 1.2) \times 10^{-13}$ , agrees well with previously known values from low-level-counting of pre-nuclear krypton. This experiment is part of a program to develop AMS for long-lived noble gas radionuclides (Kutschera *et al.* 1994). Since noble gases do not form stable negative ions, these measurements are performed with positive-ion machines.

The use of  $^{81}\text{Kr}$  ( $t_{1/2} = 2.1 \times 10^5$  a) as a valuable tracer was suggested by H. Oeschger (1987) at the Niagara-on-the-Lake AMS conference in 1987. The main interest of  $^{81}\text{Kr}$  lies in the possibility it provides for dating deep ice from the polar ice caps, and old groundwater. Due to its favorable geochemical properties it is possibly the only cosmogenic radionuclide that has the potential to become a reliable absolute chronometer for these applications. However, the extremely low concentration of  $^{81}\text{Kr}$  in these reservoirs ( $\sim 1000$  atoms per kg ice or water) demands a very high overall detection efficiency.

We have performed first AMS measurements of  $^{81}\text{Kr}$  in atmospheric krypton using the superconducting electron cyclotron resonance source coupled to the K1200 Cyclotron of Michigan State University. The main technical problem for the detection of  $^{81}\text{Kr}$  with AMS resides in the separation from the isobaric  $^{81}\text{Br}$  background. As there is still very little understanding of the source of bromine we concentrated our efforts on the separation of  $^{81}\text{Kr}$  from  $^{81}\text{Br}$  using full stripping.

After the acceleration of  $^{81}\text{Kr}^{17+}$  to 45 MeV/A in the cyclotron, the ion passed through a Be foil (18.8 mg/cm<sup>2</sup>). The fully-stripped  $^{81}\text{Kr}^{36+}$  ( $\sim 70\%$ ) is then separated from the  $^{81}\text{Br}^{35+}$  using the A1200 fragment separator. Various experimental configurations as well as support gasses and stripper foils have been tested in an attempt to optimize this method. A summary of the results for natural and neutron-activated  $^{81}\text{Kr}$  samples is given below.

Sample	$^{81}\text{Kr}/\text{Kr}$	$^{84+86}\text{Kr}^{18+}$ ( $\mu\text{A}$ )	Total $^{81}\text{Kr}$ (counts)	Background (counts)	Run time (min)	$^{81}\text{Kr}/\text{Kr}$ measured
N-act	$(2.2 \pm 0.2) \times 10^{-10}$	28.5	1044	1	22	Reference value
N-act	$(1.4 \pm 0.2) \times 10^{-11}$	17.2	373	16	219	$(1.3 \pm 0.2) \times 10^{-11}$
Natural	$(5.2 \pm 0.6) \times 10^{-13}$	19.3	34	3	405	$(5.3 \pm 1.2) \times 10^{-13}$
$^{86}\text{Kr}$	Depleted in $^{81}\text{Kr}$	21.3	7	3	413	$(6.0 \pm 4.8) \times 10^{-14}$

In order to handle small gas samples ( $\approx 3$ cc gas STP) a portable computer-controlled gas handling system was developed. The basic concept was adapted from the Oxford  $\text{CO}_2$  gas source (Bronk and Hedges 1990). Our system was designed to retain any bromine contamination from the sample gas in a heatable cold trap before the krypton sample is released into the ion source. First tests at the MSU cyclotron will be reported.

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#### IS $^{36}\text{Cl}$ FROM WEAPONS TEST FALLOUT STILL CYCLING IN THE ATMOSPHERE?

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$^{36}\text{Cl}$  concentrations in many surface waters are higher than those expected from natural production and fallout from the atmosphere. This suggests that either the existing models of  $^{36}\text{Cl}$  underestimate fallout or that there is another source of  $^{36}\text{Cl}$ . We tested the hypothesis that  $^{36}\text{Cl}$  concentrations in these systems are elevated because  $^{36}\text{Cl}$  from weapons test fallout is still cycling through the biosphere. Here we present four sets of observations to support this interpretation of the elevated concentrations of  $^{36}\text{Cl}$ .

- $^{36}\text{Cl}$  concentrations in small lakes and streams are higher than those measured in snow and rain,
- $^{36}\text{Cl}$  concentrations in shallow groundwaters (that do not contain  $^3\text{H}$ ) are lower than those measured in surface waters,
- $^{36}\text{Cl}$  concentrations in biological tissues such as tree wood, and litter, *etc.*, are much higher than values expected in natural precipitation, and
- Organic compounds collected from air contain Cl and  $^{36}\text{Cl}$ , thus providing a mechanism to recycle the isotope in different environmental pools; however, the quantity of organic  $^{36}\text{Cl}$  was very small.

These observations are consistent with the observations by Milton *et al.* (1994, NIMB) and (1995 GSA).  $^{36}\text{Cl}$  is moving through the biosphere at a slower rate than that of a non-reactive anion or water. The pulse of  $^{36}\text{Cl}$  that was introduced into the atmosphere >30 yr ago is still present in vege-

tation, organic compounds and soil and this  $^{36}\text{Cl}$  is slowly leaching into surface waters. The lag between  $^{36}\text{Cl}$  fallout and transport is not seen in data collected from ice cores because these abiotic glacial systems are dominated by the input in rain and snow and they do not have a significant biotic cycle. The behavior of  $^{36}\text{Cl}$  cannot be inferred from the distribution of  $\text{Cl}^-$  because the  $\text{Cl}^-$  mass balance is near or at steady state. The non-conservative behavior of  $^{36}\text{Cl}$  should be considered in waste management assessments and when  $^{36}\text{Cl}$  is used as a tracer in hydrologic studies when the potential for biotic cycling is significant.

### **$^{36}\text{Cl}$ AND $^{129}\text{I}$ IN TEETH AND BONES**

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Can  $^{36}\text{Cl}$  and  $^{129}\text{I}$  concentrations or atom ratios in teeth and bone be used in dosimetric studies and to date old archeological materials?

Measurements of  $^{36}\text{Cl}$  in inert biological matrices have the potential to provide a record of exposure to high neutron fluxes (*e.g.*, for the A-bomb victims and survivors). Both  $^{36}\text{Cl}$  and  $^{129}\text{I}$  could also provide information on exposures to radioiodine and volatile compounds by inhalation and/or ingestion of contaminated foodstuffs following public exposure to a release of radionuclides (*e.g.*, for people contaminated during the Chernobyl accident).

Measurements of  $^{36}\text{Cl}$  and  $^{129}\text{I}$  in archeological specimens of bone and teeth may provide a methodology to determine the age of samples that cover a range of ages from ~100k to ~200k and 15M to 75M yr before present, respectively. Samples in these age ranges are often difficult to date directly and these inert biological matrices have the potential to provide an age estimate for the actual sample.

In this presentation we will demonstrate that four of the six key issues needed to exploit these potential records of  $^{36}\text{Cl}$  and  $^{129}\text{I}$  in teeth and bone have been resolved:

1. A pyrolytic technique to extract the isotopes has been developed, tested and proven to extract the isotopes and stable analog without introducing contamination.
2. Sufficient I and Cl can be extracted to provide a sample for measurement by AMS. Stable chlorine and iodine measured in teeth and bone in concentrations range from 100 to 2000 ppm and <1 to 20 ppm, respectively, and
3.  $^{36}\text{Cl}$  and  $^{129}\text{I}$  concentrations measured in samples of teeth and bone that were exposed to known amounts of the isotopes during weapons testing have the expected concentrations of the isotopes.

We must still demonstrate that:

4. The initial concentrations of the two isotopes can be estimated accurately in order to calculate a date or subtract a background for dosimetry studies,
5. The  $^{36}\text{Cl}$  and  $^{129}\text{I}$  atom ratios that form the records in the teeth and bone are stable over the time period of interest, and
6. The estimates obtained from  $^{36}\text{Cl}$  and  $^{129}\text{I}$  are congruent with other dosimetry or age data.

Preliminary studies of these later three issues are encouraging and support the use of  $^{36}\text{Cl}$  and  $^{129}\text{I}$  in these new applications.



## **<sup>129</sup>IODINE DISTRIBUTION AROUND CHERNOBYL**

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Radio-iodine isotopes were released from the reactor during and following the Chernobyl accident. A slight excess of thyroid cancers has been reported since the accident and it is postulated that this increase in thyroid cancers results from the exposure of members of the population to the short-lived isotopes of iodine (<sup>123</sup>I to <sup>135</sup>I) during and immediately following the release. Since the longest physical half-life of these isotopes is 8 days and conditions during and immediately following the accident were difficult to assess precisely, an accurate estimate of the dose received from exposure to radio-iodine is difficult.

We report analyses of the long-lived radio-iodine isotope, <sup>129</sup>I, by accelerator mass spectrometry (AMS) performed at Chalk River Laboratories. We have used these AMS measurements to provide an independent estimate of the distribution of radio-iodine after the accident and to estimate the exposure of members of the public. Elevated concentrations of <sup>129</sup>I were measured in soils and lichen samples collected from a number of regions that received fallout from the accident. Concentrations ranged from 6 to 80 mBq kg<sup>-1</sup> of sample and the atom ratio of <sup>129</sup>I:<sup>127</sup>I ranged from 1 to 40×10<sup>-7</sup>. Concentrations in the soils and in the lichens were very similar.

The <sup>129</sup>I concentrations correlate with measurements of <sup>137</sup>Cs within an individual region. However, the relationship between <sup>129</sup>I and <sup>137</sup>Cs is different in different areas. The regional distribution of <sup>129</sup>I also correlates with that of <sup>131</sup>I. The distribution of these isotopes is consistent with the observations made during the accident that the radionuclide composition of the plume and subsequent fallout from the plume changed through out the event. This study supports the application of <sup>129</sup>I measurements to reconstruct the radio-iodine dosimetry and indicates that measurements on a variety of material including lichens, soil, foodstuffs and even the extracted teeth of individuals can provide valuable information on the exposure of individuals to radio-iodine.

## **NEW HEAVY ION LINE AT GIF-SUR-YVETTE**

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As reported at AMS-6, since 1990 we have proposed the addition of a beam line to the Gif sur Yvette AMS facility capable of analyzing isotopes over the whole periodic table. Approval of this line was given in January 1995, and construction is nearing completion. The line, very similar to that installed at IsoTrace, includes a double-focussing magnet (mass energy product of 150 Mev.amu and M/ΔM=2600) and a 45° electrostatic deflector (resolution=900). The initial application will be the analysis of <sup>129</sup>I for use both as an oceanographic tracer and in nuclear waste studies. We hope to be able to report on the first tests by the time of the meeting.

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**TRACKING BIOMASS BURNING AEROSOL: FROM THE COMBUSTION LABORATORY TO SUMMIT, GREENLAND**

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Because of its potentially profound influences on tropospheric ozone formation, visibility and climate, there is increasing interest in tracking and quantifying sources of biomass burning aerosol, especially in cases of long range transport. Among the many potential tracers for combustion aerosol,  $^{14}\text{C}$  is perhaps the most important because of its unique ability to apportion biomass and fossil carbon; and recent advances in accelerator mass spectrometry have made  $^{14}\text{C}$  "dating" of remote carbonaceous aerosol practicable, as a result of its near microgram level sensitivity. Other tracers such as potassium, ammonium, elemental and organic carbon, and specific classes of organics, such as polycyclic aromatic hydrocarbons are also widely used because of economy and/or enhanced sensitivity. Such tracers, or molecular markers, however, provide indirect evidence of biomass (or fossil) burning aerosol, so one of the most vital applications of  $^{14}\text{C}$  metrology is to assess the validity of such alternative tracers.

We shall report on a number of instances where  $^{14}\text{C}$  validation (or non-validation) is playing a critical role in regional and remote field studies, such as the application of mineral-corrected potassium to urban woodburning soot apportionment, and the application of ammonium and ion balance data to the identification of biomass burning aerosol transported to Summit Greenland, and to the development of the biomass burning record from snowpit and ice core data. The latter work is in an early stage, but already we have observed correlation between greatly enhanced trace organics with potassium, ammonium, and an ion balance variable in a Summit Greenland air particulate sample collected on 5 August 1994. Also, previously reported back trajectories for this sample showed strong transport over biomass fires in northern Canada. Complementing this work, the 1995 season at Summit provided an opportunity to look for the August 1994 biomass burning signal in snow pits, and to search for ionic, organic and isotopic biomass burning signatures in 1995 surface snow and atmospheric aerosol collected directly on quartz filters. A progress report on this work will be presented, together with vital supporting laboratory work on the combustion process, and on the production of trace organic and isotopic standards for carbonaceous aerosol research. The former, combustion studies proved extremely important in showing the relationship between trace organic aerosol composition, fuel type, and combustion stage; the latter, which includes the development of a carbonaceous particle filter standard, is vital to control the quality of carbon measurements in the remote, polar (Arctic) region where aerosol carbon is in the  $\text{mg}/\text{m}^3$  range, while its concentration in the snow and ice is of the order of  $\text{ng}/\text{g}$ .

**RADIOCARBON “DATING” OF INDIVIDUAL CHEMICAL COMPOUNDS IN ATMOSPHERIC AEROSOL: FIRST RESULTS COMPARING DIRECT ISOTOPIC AND MULTIVARIATE STATISTICAL APPORTIONMENT OF SPECIFIC POLYCYCLIC AROMATIC HYDROCARBONS**

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The ability to measure tiny amounts of radiocarbon ( $^{14}\text{C}$ ) has revolutionized our ability to apportion fossil and biogenic sources of trace carbonaceous gases and aerosols. Because of the impacts of these species on visibility, human health, and climate, accurate assessment of anthropogenic and natural sources is mandatory both for understanding the atmospheric carbon system, and for making informed decisions about that portion which may be subject to control. The chemical complexity of carbonaceous aerosol, however, presents a challenge in the interpretation of average isotopic composition. Therefore, it has long been interesting to consider ways to “date” special classes of compounds, or better still, individual compounds of particular concern. One such class comprises the polycyclic aromatic hydrocarbons (PAH), produced at trace levels (ppm) in combustion processes, such as biomass burning. The PAH hold special interest because of compound-specific genotoxicity and because of the potential for individual PAH or abundance patterns of PAH to serve a “molecular markers” for ambient aerosol sources. Prior to the work reported here, such inferences relied on indirect, multivariate statistical techniques.

The new work demonstrated, for the first time, the ability to measure  $^{14}\text{C}$  in individual, trace organic compounds in atmospheric aerosol. It utilized a special, well characterized sample collected in Washington, D.C.: “urban dust” Standard Reference Material (SRM 1649). Following extraction, individual PAH were isolated using a two-stage process involving semi-preparative liquid chromatography and preparative capillary gas chromatography, which cycles the sample stream hundreds of times through a series of traps. This process generated “datable” quantities of selected PAH which were subsequently converted to targets for  $^{14}\text{C}$  accelerator mass spectrometry, performed at Lawrence Livermore National Laboratory. Results showed considerable isotopic heterogeneity reflecting varying source contributions to different chemical fractions. The polar organic fraction, for example, had a significant biogenic component (60% fossil), whereas the aliphatic portion was almost totally derived from fossil sources (98% fossil). Among the PAH compounds dated, one—benzo(*ghi*)perylene—held special interest because previous multivariate statistical analysis of urban aerosol indicated that it might be a useful tracer for soot generated by motor vehicles. Preliminary analysis of the  $^{14}\text{C}$  data for this compound (87% fossil carbon) generally supported the statistical result.

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**RELEASE OF SEQUESTERED  $^{14}\text{C}$  DEPLETED  $\text{CO}_2$  DURING THE GROWING SEASON NEAR THE ARCTIC CIRCLE: A REGIONAL  $^{14}\text{C}$  EFFECT**

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Previously, we measured  $\Delta^{14}\text{C}$  in annual tree rings of white spruce (AD 1870–1885) from the Grand View site, Mackenzie River area of the Northwest Territory of Canada near the Arctic Circle (67°N, 130°W). We found no anomalous 11-yr  $^{14}\text{C}$  cycle as previously reported. However, we found a 2.6

$\pm 0.9\%$  ( $1\sigma$ ) depression of  $\Delta^{14}\text{C}$  relative to the Olympic Peninsula. We find a  $3.2\%$  average depression of  $\Delta^{14}\text{C}$  for single-year Douglas fir samples from the Olympic Peninsula relative to Douglas fir from the Santa Catalina Mountains near Tucson ( $32^{\circ}26'\text{N}$ ,  $110^{\circ}47'\text{W}$ ; elev. 2740 m). This implies a *ca.*  $5.8\%$  average anomaly between the Tucson mountains and Grand View site areas. Expressed in years, this would result in a *ca.* 48-yr discrepancy in ages from the two sites.

We ascribed the relative suppression of  $\Delta^{14}\text{C}$  near the Arctic Circle to the release of  $^{14}\text{C}$  depleted  $\text{CO}_2$  as thawing occurs during the May–August growing season. This is similar to the release of radon that occurs during spring thaws resulting in the tagging of air masses with anomalous amounts of radon.

In order to further study this regional effect, we extended our measurements back to AD 1861. The measured depression was a little greater than previously measured,  $3.1 \pm 1\%$  ( $1\sigma$ ) rather than  $2.6 \pm 0.9\%$  ( $1\sigma$ ), primarily resulting from an anomalous warming from AD 1867–1869 determined by dendroclimatology involving tree-ring density. The inverse correlation between  $\Delta^{14}\text{C}$  and temperature is  $r = 0.63$ , implying that *ca.* 40% of the relationship can be explained by increased thawing during warmer growing seasons. If this is correct, measurement of  $\Delta^{14}\text{C}$  in annual tree rings from Arctic regions may provide an additional paleoclimate technique.

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## **SUPERNOVA: PRIME PROBLEM OF PALEOASTROPHYSICS**

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Galileo constructed his first telescope in 1609. Prior to that, astronomers had been limited to naked-eye observations of light reaching Earth from the cosmos. Instruments have evolved to record virtually the entire electromagnetic spectrum plus protons, electrons and other particles arriving at Earth from objects distant in time and space. However, until recently the instrumental record has been limited to radiation arriving at Earth since Galileo. Cosmogenic isotope research expands the astrophysical record into the distant past limited only by the half-lives of the cosmogenic isotopes.

Cosmogenic isotopes are produced by cosmic rays and cosmic rays are of primarily galactic origin, generated by supernova explosions. If a supernova explosion were to occur near enough to the Earth, it would be observed as an increase in the cosmic ray flux, leaving a record in increased concentration of cosmogenic isotopes in natural archives. Hard  $\gamma$ -rays would arrive after the visible light and produce  $^{14}\text{C}$ . The  $8\%$   $^{14}\text{C}$  pulse recorded in tree rings in the third year after light arrived from SN 1006 AD probably signals the arrival of the hard  $\gamma$ -rays from that event. If so, the total  $\gamma$ -ray energy from SN 1006 AD is  $\sim 10^{50}$  ergs. No measurable increase in  $^{14}\text{C}$  has been observed for the Tycho and Kepler supernovae and would not be expected because of their greater distance from Earth.

The  $^{10}\text{Be}$  increase in the Vostok ice core at *ca.* 35 kyr is a candidate for arrival of a supernova shock wave at Earth. An event at approximately the same time has been observed in the GISP ice cores and in ocean sediment cores from the Gulf of California and Adriatic Sea. We believe the supernova hypothesis is correct because the increase in production rate is too great to be explained by zero geomagnetic field intensity. Geomagnetic field lines are almost vertical at Vostok and a Maunder type

minimum of such duration (~1200 yr) and magnitude is unprecedented in 10,000 yr of the Holocene  $\Delta^{14}\text{C}$  record. If so, how rare is such an event?

Our galaxy, the Milky Way, is a spiral sea level *ca.* 50,000 light years (lyr) in radius and 1000 lyr thick. It contains *ca.* 0.004 stars per (lyr)<sup>3</sup> and has a volume of *ca.*  $3.9 \times 10^{13}$  (lyr)<sup>3</sup>. Therefore, it contains *ca.*  $1.6 \times 10^{11}$  stars. Supernovae in our galaxy occur every 50 yr. Consequently, the probability of a star becoming a supernova is *ca.*  $1.3 \times 10^{-13}$  yr<sup>-1</sup> (residence time of  $7.7 \times 10^{12}$  yr). The supernova observed within the constellation Cassiopeia in 1181 AD was the furthest recorded at 28,000 lyr. The portion of the galaxy lying within a volume of that radius is *ca.* 18%. The frequency of supernovae within that volume is one every 280 yr. This compares favorably with the observed frequency of one supernova every 260 yr.

Supernova shock waves merge with the background cosmic ray flux within 100 pc (386 lyr). A sphere of that size contains *ca.*  $9.6 \times 10^5$  stars and the expected supernova frequency is one per 8 my. This is approximate; we have assumed a uniform galaxy. Nevertheless, such events must be very rare! Yet, according to astronomers, we live within the swept out "bubble" of a supernova shock wave that passed through the solar system. We may have evidence for a real but rare event. The remnant is probably a nebula. Which one, the Veil Nebula in Cygnus, the SN remnant within the North Polar Spur, Geminga, others?

Obviously more work needs to be done, *e.g.*, more detailed <sup>10</sup>Be measurements in cores with minor bioturbation and fast sedimentation rates. Measurement of other isotopes such as <sup>26</sup>Al, <sup>36</sup>Cl, <sup>41</sup>Ca and <sup>129</sup>I would be desirable. There should be a search for *in-situ* production of <sup>14</sup>C and the deposition of nitrates in polar ice. What were the paleoenvironmental consequences involving climate and space environment and what was the affect on organisms living within that millennium?

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## TEAMS DEPTH PROFILES IN SEMICONDUCTORS

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Accelerator mass spectrometry (AMS) is routinely used to measure abundance ratios of long-lived radioisotopes such as <sup>36</sup>Cl, <sup>14</sup>C and <sup>129</sup>I to their stable isotopes at levels as low as  $1 \times 10^{-15}$ . Secondary ion mass spectrometry (SIMS) is one of the most sensitive techniques for the determination of impurity depth profiles in semiconductors. Trace element accelerator mass spectrometry (TEAMS) is the combination of these two techniques, applied to the measurement of very low levels of stable elements in a matrix that may be quite different from the element being detected. TEAMS offers the possibility of detection limits of the order of tens of ppt for certain impurities in silicon, which is substantially better than SIMS. In general, TEAMS data is subject to the same constants as SIMS, the big improvement arising from the elimination of molecular interferences that bedevil SIMS. The Ion Beam Modification and Analysis Laboratory at the University of North Texas has a dedicated facility for TEAMS measurements. A detailed description of the laboratory and TEAMS apparatus will be presented along with recent TEAMS depth profiles from a variety of implants in semiconductors.

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## **A POSSIBLE APPLICATION OF NEW DISCOVERIES IN VACUUM HIGH-VOLTAGE INSULATION**

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An investigation of vacuum high-voltage insulation processes has been carried out at the Tandem Accelerator Superconducting Cyclotron (TASCC) facility with the goal of improved operation of the cyclotron electrostatic deflector. This research program has produced an improved understanding of the underlying processes of electrical breakdown in vacuum. Electric fields as high as 90 MV/m at a 1-mm gap and 50 MV/m at a 4-mm gap have been reached with no measurable field emission. The control of field emission is key to the realization of an electrostatic accelerator designed for vacuum insulation instead of gas. This paper will present an approach to a new accelerator design inspired by our research program.

## ***IN-SITU* AMS DETERMINATION OF Re-Os ISOCHRON IN IIA IRON METEORITES**

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Using two electrostatic plate deflectors, we have developed an isotope switching system for the IsoTrace AMS system. It was applied to the *in-situ* determination of Re-Os isochrons for the Ni-Fe phases of a set of iron meteorites with emphasis on group IIA. The method requires no chemical sample treatment and permits analysis of the Re-Os systematics of materials at sub-mm scales. The fitted isochron of IIA irons corresponds to an age of  $4.70 \pm 0.31$  Ga, consistent with the latest reported IIA ages of  $4.584 \pm 0.043$  Ga (Morgan *et al.* 1995) and  $4.61 \pm 0.01$  Ga (Shen, Papanastassiou and Wasserburg 1995), obtained by conventional whole-rock methods. The detection limits of Re and Os are estimated to be 54 ppb and 0.5 ppb, respectively. It is shown that *in-situ* AMS has the capability to obtain an internal Re-Os isochron in iron meteorites. It also provides a convenient and sensitive tool to study Re-Os systematics in broad geological applications.

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## EVIDENCE OF ANOMALOUS $^{107}\text{Ag}$ AND $^{109}\text{Ag}$ COMPOSITION IN IRON METEORITES

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It has been reported that excessive  $^{107}\text{Ag}$  in a wide range of iron meteorites is correlated with the Pd concentrations. Anomalous  $^{107}\text{Ag}$  was attributed to the *in-situ* decay of  $^{107}\text{Pd}$  still alive in the early solar system (Wasserburg 1985). We applied the *in-situ* AMS method to determine  $^{107}\text{Ag}$  and  $^{109}\text{Ag}$  compositions in several groups of iron meteorites and observed extreme variations of  $^{107}\text{Ag}$  and  $^{109}\text{Ag}$  count rates from those samples as a function of measurement sequence. In contrast, no significant variation in the counting rate of  $^{108}\text{Pd}$  was observed. The origin of the inclusions responsible for these variations are not understood. However, in one interpretation, the observed results could be attributed to the preservation of pre-solar grains at sub- $\mu\text{m}$  scales in iron meteorites. New AMS methods of measuring  $^{107}\text{Ag}$  and  $^{109}\text{Ag}$  simultaneously will be discussed.

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## A SERVER-BASED CODE FOR *IN-SITU*-PRODUCED NUCLIDES THAT INCORPORATES IRREGULAR GEOMETRIES

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The application of *in-situ*-produced radionuclide dating of rock surfaces requires complex calculations and the incorporation of a few to as many as 100 pieces of data for each sample. Full neutron transport calculations, sometimes needed for  $^{36}\text{Cl}$ , can take from a few hours to a day or more on high-speed computers. Results need to be presented in terms of both exposure ages and erosion rates. To expedite this, a versatile computer code, radionuclide *in-situ*-produced cosmogenic-nuclide history (RICH), has been developed. This code is available to all scientists on the internet.

RICH is designed to accept data, with uncertainties, for any parameter that can affect production rate. Production rates are scaled for latitude, altitude, geometric shielding, depth, burial, snow cover, and rock composition. RICH will solve for the erosion rate, which (for now) is assumed constant. Complex, finite sampling surfaces are being modeled by incorporating Monte Carlo neutron, proton, and muon transport codes with an appropriate seed particle flux tuned to the site location. We have plans to account for geomagnetic variations, tectonic uplift, and multiple stepwise erosion.

Because development of cosmogenic rock-exposure dating requires constant re-evaluation of old data in the light of new refinements, RICH accepts all parameters, such as production rates, as input. Alternatively, most parameters can remain as defaults for quick estimates prior to sampling. RICH can combine data from several different isotopes or from two different  $^{36}\text{Cl}$  mineral separates within the same run. RICH propagates uncertainties for all data and parameters. RICH runs as a batch process on an Internet-accessible Digital Alpha workstation, and will soon support a World Wide Web on-line interface for entering data, in addition to the current e-mail interface.

### ISOLATION OF INDIVIDUAL ORGANIC COMPOUNDS FOR AMS RADIOCARBON ANALYSIS: A NOVEL APPROACH

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The ability to measure natural levels of radiocarbon in individual organic compounds isolated from geologic samples adds a new dimension to the types of studies one can conduct. Successful application of this technique requires the collection of sufficient quantities of compounds usually present in very low concentrations in complex matrices and the ability to measure small samples on the AMS. In this study, isolation of compounds containing 10–200  $\mu\text{g}$  C is accomplished using preparative capillary gas chromatography (PCGC). PCGC allows one to collect the same peak from repeated injections (typically 20–100) in order to isolate sufficient material for an AMS analysis. The isolated compounds are transferred to a combustion tube with a solvent, the solvent is evaporated, and then the sample is treated like a small AMS sample. We have found it is best to isolate at least 25  $\mu\text{g}$  C and that it can be difficult to remove all of the solvent for some compounds. We tested this technique in a pilot study (Eglinton *et al.* in press) and found that we can accurately date individual compounds. It is best to isolate at least 25  $\mu\text{g}$  C and, for some compounds, extra care must be taken to remove all the solvent. We are now applying the technique to study the origin of organic matter in surficial marine sediments. Initial results from the Black and Arabian Seas indicate that biomarker compounds from distinct sources have significantly different ages. For example, in Arabian Sea sediments, markers for diatoms (highly branched isoprenoid alkenes) were very young (~250 yr) compared to the saturated hydrocarbons (~8000–10,000 yr). We will describe the technique in detail, including the analysis of samples on the AMS. We will also discuss research areas where this technique may be useful.

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### THE MICROSTRIP GAS CHAMBER FOR USE IN GAS-FILLED MAGNETS

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Gas-filled magnets (GFM) are becoming popular as isobar separators in AMS systems. They are useful for measurement of  $^{36}\text{Cl}$  where  $^{36}\text{S}$  is difficult to separate with 5–6 MV tandems and where samples are small or chemical preparation is not adequate. GFM are essential for heavier nuclides



such as  $^{59}\text{Ni}$ ,  $^{60}\text{Fe}$ , and  $^{53}\text{Mn}$  where isobars are more abundant and more difficult to separate. Even at the highest energies used for AMS, the GFM is used primarily as a count-rate reducer and much of the isobar separation is performed in the gas ionization detector that follows the GFM. It is clear that the GFM as presently used will not make isobar separation easy at low energies without advances in performance.

The radius of curvature of heavy ions in a GFM is proportional to  $(ME)^{1/2}/q$  where  $M$  is the mass and  $E$  the kinetic energy. Because of the statistical charge-changing process in the gas, the average charge state  $q$  depends on  $Z$ , the atomic number. Thus ions of different  $Z$  leave the magnet at different locations, which allows one to stop the unwanted isobar from entering the detector. The isobar selectivity is limited by multiple small angle scattering in the entrance foil and in the gas in the first part of the magnet. The isobar separation would be greatly improved by measuring the radius of curvature of the ions at several places inside the magnet. This requires a position-sensitive detector that will fit in the small space inside the magnet chamber and one that can handle high counting rates.

Micro-strip gas chambers (MSGC) are being developed for low-ionizing particles at high counting rates for experiments in high energy physics (see Proceedings of the International Workshop on Micro-Strip Gas Chambers, Legnaro, Italy, 13–14 October 1994). They consist of narrow (cathode) and wide (anode) strips placed on a glass substrate by lithography techniques, typically with a 1 mm pitch. Ionization takes place only near the cathode and positive ions are removed quickly since the anode is very close to the cathode. This allows high counting rates even for highly ionizing particles, potentially making the MSGC a good position detector for use inside a GFM. There is little experience with the use of the MSGC with heavy ions. Results of some tests with beams will be presented.

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## STATUS AND PLANS FOR THE PRIME LAB AMS FACILITY

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The Purdue Rare Isotope Measurement Laboratory is the National Science Foundation facility for measurement of the nuclides  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ , and  $^{129}\text{I}$ .  $^{14}\text{C}$  is also measured, but at low priority since the NSF supports two other facilities to measure this nuclide. The AMS system is based on an upgraded FN (8 MV) tandem accelerator (Mueller *et al.*, this conference). The ion source (Miller *et al.*, this conference) is a spherical-ionizer cesium-sputter source with a custom 8-sample wheel. Isotope ratios are obtained by cycling the injector and analyzer magnets (Perry *et al.*, this conference), with plans for fast cycling (Purser *et al.*, AMS-6). The multi-plate gas ionization detector and data acquisition system have special features for separation of  $^{36}\text{Cl}$  and  $^{36}\text{S}$  (Knies *et al.*, AMS-6). The data-acquisition system has been automated; samples are measured overnight without need for operators. Sample results are available to users on the World Wide Web as soon as they are measured. The existing on-line sample database is being upgraded to Oracle (X. Ma, this conference). Quality assurance and quality control procedures are being implemented.

PRIME Lab chemistry operations offers a physical and chemical sample preparation service for the local and international AMS community. Construction of two new sample preparation laboratories

(Vogt *et al.*, AMS-6) is complete and preparation techniques for the nuclides listed have been developed for a wide variety of matrices, including  $^{10}\text{Be}$ ,  $^{36}\text{Cl}$  and  $^{129}\text{I}$  in water,  $^{26}\text{Al}$  and  $^{10}\text{Be}$  in quartz,  $^{10}\text{Be}$  in soils and  $^{36}\text{Cl}$  in various rock mineral fractions. AMS of all nuclides listed has been demonstrated with the exception of  $^{41}\text{Ca}$ ; work on  $^{41}\text{Ca}$  detection using  $\text{CaH}_2$  and  $\text{CaF}_2$  is in progress.

We have constructed a graphite production line and demonstrated 1% precision for  $^{14}\text{C}/^{13}\text{C}$  ratios; capability for this nuclide is being developed primarily for local projects and for projects that involve measurement of other AMS nuclides at PRIME Lab. In 1995 we measured a total of 1265 research samples for five nuclides:  $^{10}\text{Be}$ , 162;  $^{14}\text{C}$ , 31;  $^{26}\text{Al}$ , 427;  $^{36}\text{Cl}$ , 577;  $^{129}\text{I}$ , 68. Of these, 260 were chemically prepared at PRIME Lab. We loaded a total of 3950 samples (including tests, standards, and blanks) into cathodes in 1995.

There are over 125 users of our facility involved in 20 internal and 160 external (Sharma *et al.*, this conference) projects. Purdue-based goals in earth and planetary sciences are to calibrate and develop methods for using cosmogenic radionuclides to study the history of rocks, soils, meteorites, and groundwater through improvements in sample collection methods, chemical separation techniques and theoretical models. Applications reported here include glacial erosion of bedrock, the glacial history of Indiana, rock erosion rates, volcanic and tectonic history of the Andes, Antarctic meteorites and the Mocs meteorite strewnfield. Purdue-based goals in the biomedical sciences are to develop methods for tracing aluminum, calcium and carbon-containing compounds in living systems; applications reported here include aluminum uptake in rats, aluminum-containing adjuvants and cholesterol.

#### STABLE TRACE ELEMENT AMS AT THE PSI/ETH FACILITY IN ZURICH

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The limits of conventional Secondary Ion Mass Spectrometry (SIMS) for the detection of stable trace elements in ultrapure materials can be extended with Accelerator Mass Spectrometry (AMS), since it strongly suppresses all molecular background. Based on the results of trace element analysis tests (Döbeli *et al.* 1994) obtained with the existing AMS setup (developed for radioisotope detection) a new sputtering chamber has been built to fulfil the special requirements of materials analysis (Ender, in press). The major differences between the old and the new sputtering chamber are a less intense but mass analyzed and well focused Cs sputter beam, a better vacuum (~10–9 mbar) and an extraction system which suppresses the contamination of the sample by the sputtering process.

The Cs sputter gun of the new setup is a commercial SIMS product. It is equipped with an ExB filter and a 1° beam bend for neutral particle suppression, resulting in a pure Cs ion beam. The ion energy is variable between 1 and 15 keV. With different apertures it is possible to select a range of beam focus sizes (2–150 mm) and the corresponding ion currents (40 pA–500 nA). These parameters can thus be optimized for different applications such as bulk analysis, depth profiling and imaging.

The secondary ion extraction geometry is adapted to the ion optical requirements of the existing accelerator mass spectrometer. It is realized with a cylindrical acceleration lens with four electrodes, accelerating the ions to 40 keV. For the injection into the existing beamline a retractable, spherical electrostatic deflector is used.

In order to keep contamination of the extracted ion beam as low as possible, the entire environment of the target is plated with a 20-mm-thick layer of pure gold. This reduces the background level of

any element but gold by several orders of magnitude. The setup features high extraction and transmission efficiencies, but low contamination from sputtering and redeposition.

The detection system includes a Faraday cup and a channelplate detector on the low energy side and a Faraday cup, a channelplate and a gas ionization detector on the high-energy side of the accelerator. This allows the measurement of the matrix element current as well as single event counting of the trace element of interest. The mass to be injected and to be analyzed as well as the charge state and the detector type to be used are selectable *via* computer control.

The complete setup (Cs gun, sputtering chamber, detection system, computer control, data acquisition and special design aspects) as well as first test results will be presented and compared with the experiments performed with a standard AMS setup.

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## COSMOGENIC CHLORINE-36 PRODUCTION RATES FROM POTASSIUM

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We are calibrating the *in-situ* production rate of <sup>36</sup>Cl from potassium in surface rocks using the following strategy: 1) Work with separated minerals (*e.g.*, K-feldspar) to maximize both target element abundances (enhancing analytical sensitivity) and K/Cl ratios (enhancing contributions from spallation relative to slow neutron capture); 2) suppress <sup>36</sup>Cl measurement errors and reduce dependence on the accuracy of Cl analyses by adding Cl carrier to samples; 3) determine in individual samples the <sup>36</sup>Cl contribution from slow neutron capture by <sup>35</sup>Cl; and 4) use deep sub-surface samples to calibrate the muon reaction.

To date, only surface samples of K-feldspar have been measured, precluding a breakdown of total production from K ( $P_K$ ) into component reactions, but by sampling from geographically distinct sites we can reasonably estimate global surface production rates from potassium.

Assuming  $t_{exp} = 13.1$  ka, samples from the Sierra Nevada give  $P_K = 1460 \pm 88$  atom (gK)<sup>-1</sup> a<sup>-1</sup> at 3000 m and  $\lambda_{eff} = 37.4^\circ$ . Concordant results are obtained from samples at 4 sites, at elevations between 3000 and 3600 m and with chloride concentration from 10–310 ppm. Reactions on K account for 76–99% of production in the samples. Consistency between samples with low and high Cl contents is obtained by assuming a surface neutron capture rate of 2450 n g<sup>-1</sup> a<sup>-1</sup> at 3000 m and  $\lambda_{eff} = 37.4^\circ$  (close to the scaled value of Zreda *et al.* 1991). Assuming 5% of K production at sea level is due to muon capture, these data give  $P_K = 180 \pm 11$  atom (gK)<sup>-1</sup> a<sup>-1</sup> at sea level and high latitude.

Samples from pavements exposed by retreat of Loch Lomond (Younger Dryas) Stage ice ( $t_{exp} = 11.5 \pm 0.3$  ka) in Scotland give  $P_K = 313 \pm 25$  atom (gK)<sup>-1</sup> a<sup>-1</sup> at 520 m and 58.5°N. Neutron capture corrections are 18–20% for these samples. Again assuming 5% of total K production due to muon capture, these data give  $P_K = 189 \pm 15$  atom (gK)<sup>-1</sup> a<sup>-1</sup> at sea-level and high latitude, in excellent agreement with results from the Sierra Nevada.

Two samples of K-feldspar from the Trans-Antarctic Mountains (77.5°S), apparently saturated with  $^{36}\text{Cl}$ , give production rates of  $1350 \pm 50 \text{ atom (gK)}^{-1} \text{ a}^{-1}$  at 2050 m and  $1230 \pm 40 \text{ atom (gK)}^{-1} \text{ a}^{-1}$  at 2000 m, with negligible (<2%) corrections for neutron capture on  $^{35}\text{Cl}$ . Equivalent rates at sea level, scaled as above are  $P_K = 239 \pm 10$  and  $227 \pm 8 \text{ atom (gK)}^{-1} \text{ a}^{-1}$ . The samples have been measured repeatedly to confirm that the 25% discrepancy with the scaled values from Scotland and the Sierra Nevada is real and must be accounted for either by the scaling procedure, or secular variation in the cosmic ray flux.

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## PRE-ANTHROPOGENIC $^{129}\text{I}$ IN THE MARINE SYSTEM: OBSERVATIONS AND APPLICATIONS

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$^{129}\text{I}$ , the only long-lived radioisotope of iodine, is of interest because of its potential applications for tracing and dating of processes in the age range between 5 and 80 Ma. The biophilic nature of iodine and the low abundance of this element in rock-forming minerals sets this system apart from other isotopic systems. The main production modes for  $^{129}\text{I}$  are the interaction of cosmic rays with Xe isotopes in the atmosphere and the spontaneous fission of  $^{238}\text{U}$  in the crust. In order to apply this system successfully, the input function and behavior of this isotope in the marine system has to be understood. We report here results from the determination of  $^{129}\text{I}/\text{I}$  ratios in marine sediments, deep waters and marine organisms which were undertaken in order to determine the pre-anthropogenic signal of this isotopic system in the oceans.

The results suggest a pre-anthropogenic  $^{129}\text{I}/\text{I}$  ratio of  $1500 \times 10^{-15}$ , in good agreement with earlier studies done in marine sediments. However, variation in the ratios in most of the sediment cores was found to be much larger than the experimental error. The most likely interpretation of this variation is related to the presence of a fossil carbon component in the marine sediments. Because this fossil carbon does not participate in the active carbon cycle, iodine bound to it will preserve the age of its original formation. The oldest iodine was found in a core off the Oregon coast with a ratio of  $354 \times 10^{-15}$ , suggesting Miocene age for the fossil carbon in this core.

Our results indicate that iodine is present in two major forms in the marine system: a labile constituent which is readily available for incorporation into organic organisms and a component which is bound to refractory carbon and does not participate in the iodine recycling in the oceans. Our results show that the labile component is well mixed in the marine reservoir and has an input ratio of  $1500 \times 10^{-15}$ . Since organisms will take up the labile component, this ratio is to be used for the calculation of ages using  $^{129}\text{I}$  in hydrologic systems or in organic material. The association of  $^{129}\text{I}$  with fossil carbon opens up the possibility of determining age and amount of refractory carbon, an important question for the understanding of the global carbon cycle.

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## COSMOGENIC-RADIONUCLIDE PROFILE OF THE MOCS METEORITE STREWNFIELD

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Cosmic-ray produced nuclides were measured in samples from eight pieces of the L5-6 chondrite Mocs from known locations in the strewnfield (Graham *et al.* 1985; Koch 1882). We measured  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in the bulk phase along with  $^{36}\text{Cl}$  in both the metal and silicate phases.  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$  were chemically separated as described by Vogt and Herpers (1988). All samples were measured at the Purdue accelerator mass spectroscopy facility (PRIME Lab) (Elmore *et al.* 1994).

Measurements of cosmogenic radionuclides in individual meteorites have been used to provide information on the geometry of meteoroids (Graf *et al.* 1990). However, no similar data exist on the strewnfield of a meteorite shower. It is well known that larger fragments tend to travel farther and impact at the far-end of the strewnfield. As a working hypothesis, we might expect a systematic variation of cosmogenic radionuclide levels with location of fragments in the strewnfield. While further measurements will be required to fully test this hypothesis, current results do suggest a  $>2\pi$  irradiation for the Mocs meteoroid which was  $<1$  m in radius.

Depth of samples from the surface of the parent meteoroid can be estimated from  $^{10}\text{Be}$  and  $^{26}\text{Al}$  measurements.  $^{10}\text{Be}$  values in this work ranged from  $20.5 \pm 0.5$  to  $23.7 \pm 0.4$  dpm/kg. Previous measurements of noble gases resulted in  $^{22}\text{Ne}/^{21}\text{Ne}$  ratios of 1.04–1.10 (Herzog 1973). Based on relationship of  $^{10}\text{Be}$  activities to  $^{22}\text{Ne}/^{21}\text{Ne}$  ratios, all samples probably originated from depths of 60 to 80 cm (Graf, Baur and Signer 1990). Activities of  $^{26}\text{Al}$  measured in this work also varied little, ranging from  $66 \pm 2$  dpm/kg to  $74 \pm 4$  dpm/kg. These values agree well with previously published results, which range from  $69 \pm 2$  to  $74 \pm 3$  dpm/kg (Cameron and Top 1975) This also suggests that these samples originated from a depth of approximately 70 cm in the parent meteoroid (Reedy 1987). Activities of  $^{36}\text{Cl}$  showed slightly more variation than either  $^{10}\text{Be}$  or  $^{26}\text{Al}$ , with metal phase  $^{36}\text{Cl}$  ranging from  $13.1 \pm 0.2$  to  $16.5 \pm 0.4$  dpm/kg and preliminary  $^{36}\text{Cl}$  in the silicate phase ranging from  $7.5 \pm 0.1$  to  $12.1 \pm 0.2$  dpm/kg.

Despite large uncertainties, the three cosmogenic nuclides exhibit an interesting trend, with six of the eight samples apparently defining a linear relationship between  $^{10}\text{Be}/^{26}\text{Al}$  and  $^{36}\text{Cl}/^{26}\text{Al}$ . The only two outliers of this trend likely came from a secondary strewnfield superimposed on the main strewnfield. These data hint at a one-stage irradiation for most Mocs samples with a more complicated history for some.

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**AMS OF THE PLANETARY ELEMENTS**

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The detection of long-lived plutonium and neptunium isotopes by AMS at high sensitivity has been demonstrated with the 14UD accelerator at the Australian National University. Possible applications include nuclear safeguards and waste disposal issues, the biogeochemical behavior of these elements following release into the environment, and the biochemistry of plutonium. Briefly, the methodology is as follows: the plutonium or neptunium is dispersed in iron oxide and PuO<sup>-</sup> or NpO<sup>-</sup> ions extracted from a SNICS high-intensity source. Gas stripping is employed, and to date the 7<sup>+</sup> charge state has been selected for analysis. The choice of accelerating voltage is dictated by the maximum mass-energy product (135 MeV·amu) of the high-energy beam transport system, which for Pu<sup>7+</sup> ions corresponds to an energy of 28 MeV and a terminal voltage of 3.5 MV. Pu and Np ions are detected in a propane-filled ionization detector in which the electric field is parallel to the direction of the incoming ions. An energy resolution of 3.1% is achieved for 28 MeV actinide ions which is more than adequate to resolve the Pu and Np from all charge-exchanged ions with the exception of other actinides, principally <sup>238</sup>U.

A number of tests of the system have been performed and show that it is capable of quantitative measurement of isotope ratios for these very heavy elements. Backgrounds are very low. First measurements for projects to study the distribution of Np in pore waters from mud flats in the vicinity of the Sellafield reprocessing plant in Cumbria and to measure the Pu levels in people living near the plant are underway. Preliminary results will be presented. In addition, the prospects for improvements in performance will be discussed.

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**<sup>10</sup>Be MEASUREMENTS AT THE ANTARES AMS FACILITY**

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Measurements of <sup>10</sup>Be have commenced at the ANTARES AMS facility. First results compare very well with that obtained at other AMS laboratories routinely measuring <sup>10</sup>Be. Plans are to increase the <sup>10</sup>Be measurement program through 1996 related to projects in the Earth Sciences (geomorphology, subduction and ice cores).

A conventional gas ionization detector (using P10) preceded by an Ar absorber cell with Havar windows, a TV of 5.6 MV and selection of the 3<sup>+</sup> charge state were used to identify <sup>10</sup>Be. Particle transmission (using C stripper foils) was 15–20% and future work will employ our new gas stripper. The injector magnet was bounced at 2 Hz (50 msec <sup>9</sup>Be, 450 msec <sup>10</sup>Be) with <sup>10</sup>Be/<sup>9</sup>Be ratios presented at 2–3-min intervals.

Blank BeO produced from dissolved beryl consistently gave 10/9 ratios of  $1 \times 10^{-14}$  while that from our commercial BeO ranged from  $5-7 \times 10^{-14}$ . Four standards (3 kindly supplied by S. Vogt from PRIME lab and one from University of Pennsylvania), ranging from  $1 \times 10^{-11}$  to  $1 \times 10^{-8}$ , were mea-

sured sequentially, each to within 10% of their respective nominal values. Background subtraction as a function of absorber boron rates will be used to correct measured ratios. Alternating extended measurement of a high level standard with the best blank showed a short term ion-source memory of  $\sim 2 \times 10^{-4}$ , reducing by a factor of 5 after 60 min.

Samples from 4 volcanic sites related to subduction projects were prepared at the University of Queensland using known methods for boron elimination. Ion source currents were  $\sim 2 \mu\text{A}$  (compared to 4–6  $\mu\text{A}$  from blanks and standards) but two samples gave unacceptable boron rates. Blanks and standards showed negligible boron. Two meteorite samples, Tabor and Murray, which had previously been analyzed at University of Pennsylvania, were measured at ANTARES ( $18.1 \pm 1.3$  and  $10.4 \pm 1.0$  dpm/kg) in excellent agreement ( $\pm 5\%$ ) with the University of Pennsylvania data.

### **$^{110}\text{Be}$ , $^{26}\text{Al}$ AND $^{36}\text{Cl}$ RESEARCH TOPICS IN EARTH SCIENCES AT ANTARES**

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In parallel with a comprehensive  $^{14}\text{C}$  measurement program, over the past two years, routine measurement of  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$  and more recently  $^{10}\text{Be}$ , has been demonstrated at ANTARES. The research program at ANTARES dealing with these radioisotopes is focussed on the Earth Sciences, largely in the fields of geomorphology and volcanism.

Six projects using *in-situ* production of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$  are in progress. Sample collection has been completed for all but one, and sample pretreatment and processing has commenced. These projects deal with: 1) the glacial history of the Prince Charles Mountains, Antarctica, using bedrock and boulder samples to estimate both exposure ages and uplift rates (with Barrie McKelvey, University of New England); 2) timing and extent of deglaciation of the Pleistocene ice-cap on the Central Plateau of Tasmania using  $^{36}\text{Cl}$  in moraine boulders and  $^{14}\text{C}$  in sub-algal lake deposits (David Hannon, University of Tasmania); 3) measurement of  $^{10}\text{Be}$  in deep sand/sandstone regoliths along the coastal plains of Western Australia to distinguish between possible formation processes by either laterization or deposition (David Newsome, Murdoch University, WA); 4) exposure dating of surface formations exhibiting evidence of paleosismic events such as avalanches, slab fractures and pop-ups throughout central Australia; 5) dating the advance of weathering fronts in the formation of stepped granite bornhardts and of pediment surfaces with associated scarp bevels (Flinders Ranges) (Raul Twidale, University of Adelaide, SA); and 6) date ages of deposited and eroded impact of catastrophic wave trains on the southern Australian coastline using large boulders and wave-eroded bedrock (with Robert Young, University of Wollongong).

Rock crushing, mineral selection and pretreatment have been carried out at two of the above universities (UNE and Tasmania), with rock and/or quartz dissolution to be performed at ANTARES. A total of  $\sim 100$  field samples are involved and new *in-situ* projects are being presently evaluated.

Five projects are underway using  $^{10}\text{Be}$  to study subduction processes with samples from various island-arc volcanic zones, including Indonesia, Tonga, New Zealand and Japan. All samples are well characterized with respect to geochemistry and petrology, and most have available Sr, Nd and Pb systematics. Sample processing (by Tony Ewart) has commenced at the University of Queensland. A total of 55 sediment/volcanic samples will be analyzed (by John Foden, Janet Hergt, David Gust and Ian Smith at the Universities of Adelaide, Melbourne, Queensland and Auckland, respectively).

## SOLAR AND GEOMAGNETIC EFFECTS ON THE CONCENTRATION OF $^{10}\text{Be}$ AND $^{36}\text{Cl}$ IN THE GISP2 ICE CORE

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$^{10}\text{Be}$  and  $^{36}\text{Cl}$  are produced in the atmosphere by the interaction of high-energy cosmic ray primaries and secondaries with atmospheric gases. Both solar and terrestrial magnetic fields can influence the production rates of these radionuclides in the Earth's atmosphere. Changes in atmospheric circulation and in snow accumulation rate can in turn affect the radionuclide concentration in polar ice samples. In order to study these effects, we measured a continuous profile of  $^{10}\text{Be}$  and  $^{36}\text{Cl}$  concentrations in the GISP2 ice core, which spans the time period from the present to beyond 100 kyr BP.  $^{10}\text{Be}$  concentrations were on the order of a few times  $10^4$  at/g ice and  $^{36}\text{Cl}$  concentrations about a factor of 5 less. We observed a large decrease in radionuclide concentration, about a factor of two, at the end of the Younger Dryas. Fluxes of  $^{10}\text{Be}$  and  $^{36}\text{Cl}$  calculated using the GISP2 accumulation rates, exhibited little or no change in concentration correlated with variations in the geomagnetic field such as are observed in the tree-ring  $^{14}\text{C}$  record. We do, however, detect fluctuations on a centennial time scale. These fluctuations are interpreted as resulting from variations in the solar magnetic field associated with quiet sun periods similar to the Maunder Minimum.

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## IN-VIVO ABSORPTION OF ALUMINUM-CONTAINING ADJUVANTS USING $^{26}\text{Al}$

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Aluminum-containing adjuvants are used in vaccines because they enhance antibody production. Interest in adjuvants has increased due to the availability of biotechnology-generated antigens. Aluminum-containing adjuvants are important because they are the only type approved for use by the Food and Drug Administration, and increased knowledge about adjuvants will help facilitate the production of more effective vaccines. Measurement of  $^{26}\text{Al}$  by AMS offers the first opportunity to demonstrate that adjuvants are removed from the site of injection by dissolution in the interstitial fluid. Furthermore, the relative rates of *in-vivo* absorption of crystalline aluminum hydroxide and amorphous aluminum phosphate adjuvants can be determined.

Procedures for precipitating small quantities of two  $^{26}\text{Al}$  enriched adjuvants, aluminum hydroxide and aluminum phosphate, were developed and tested to insure that the  $^{26}\text{Al}$  labeled adjuvants have the same chemical and physical properties as commercial versions. These adjuvants were then injected intramuscularly (IM) in New Zealand White rabbits (2 rabbits/adjuvant). One rabbit received an intravenous (IV) injection of  $^{26}\text{Al}$  citrate, and an additional rabbit was used as a cross-contamination monitor. Each rabbit received a total of 0.85 mg Al, which is the standard amount contained in a vaccination. Blood samples were obtained periodically for 28 days, and tissue samples were collected after day 28. Samples were then analyzed using AMS.



Data was analyzed using model independent analysis. The mean residence time (MRT) of  $^{26}\text{Al}$  in the rabbit with the IV dose was  $\sim 1.4$  hr. The normalized area under the blood concentration curve (AUC) from  $t=0$  to  $t=28$  days was found to be  $3.3 \times 10^{-4} \text{ g}^{-1} \text{ h}$  for the aluminum hydroxide,  $9.4 \times 10^{-4} \text{ g}^{-1} \text{ h}$  for the aluminum phosphate and  $1.9 \times 10^{-3} \text{ g}^{-1} \text{ h}$  for the IV dose. This indicates that, by day 28, 3 $\times$  as much aluminum phosphate had been absorbed as aluminum hydroxide. Blood concentration of  $^{26}\text{Al}$  for each of the rabbits receiving an adjuvant had not reached a terminal phase by day 28, so it is difficult to determine the MRT of the adjuvants. However, it is clear that the aluminum hydroxide has a bi-modal absorption phase where the MRT of the first phase is  $<50$  hr, and the MRT of the second phase is much longer. This difference in the dissolution profile of the two adjuvants has implications for the release profile of the absorbed antigen in vaccines.

The increase in the aluminum plasma concentration from the 0.85 mg dose was approximately 1 ng/ml which was masked by the background aluminum plasma concentration.

Distribution of  $^{26}\text{Al}$  among tissues for both adjuvants was bone > kidney > spleen > liver > heart > lymph > brain.

For each tissue, retention of  $^{26}\text{Al}$  was greater for the aluminum phosphate adjuvant than the aluminum hydroxide adjuvant, which reflects the greater dissolution of the aluminum phosphate.

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## THE RECONSTRUCTION OF ATMOSPHERIC TRACE GAS COMPOSITION FROM ANTARCTIC ICE CORES

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High-precision measurements of carbon dioxide, methane and other species over the last 1000 yr have been carried out on air extracted from bubbles in coastal Antarctic ice from Law Dome. Unique features of these records are a consequence of a very high accumulation rate coupled with low annual temperatures, and include the overlap of the most recent trapped air with direct atmospheric measurements plus high time resolution.

The reconstruction of an atmospheric trace gas history from the ice core measurements is not straightforward and requires detailed knowledge of the effects of diffusion in the firn layer overlying the ice, and in the bubble close-off process. The corrections for diffusion effects are second-order for typical trace gas concentration precisions but become critical for higher-precision measurements such as the stable isotope ratios of  $\text{CO}_2$  and  $\text{CH}_4$ . In all cases the histories are smoothed by diffusion and trapping effects.

AMS dating of  $\text{CO}_2$  has played a key role in confirming the magnitude of diffusion and trapping corrections, and the degree of smoothing. AMS measurements on "bomb-spike"  $^{14}\text{CO}_2$  in air from firn and ice are used as an independent check on the output of a firn diffusion and trapping model, which is initially calibrated with firn measurements for trace gas concentrations representing a range of diffusion coefficients and atmospheric growth rates.

The importance of the AMS-validated model is demonstrated with regard to atmospheric histories of  $\delta^{13}\text{CO}_2$  and  $\delta^{13}\text{CH}_4$ . Long isotope time series of recent measurements obtained from the Cape Grim Air Archive, a suite of samples of marine air collected in stainless steel containers since 1978, provide a unique opportunity to further verify the corrections.

Biogeochemical implications of the isotopic records are briefly discussed.

## **THE ROLES OF $^{13}\text{C}$ AND $^{14}\text{C}$ IN UNRAVELING THE GLOBAL ATMOSPHERE CARBON BUDGET**

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High-precision monitoring in the southern hemisphere of atmospheric carbon dioxide and its isotopic composition has revealed features not easily accommodated by existing models. The importance of understanding subtle changes in southern hemisphere atmospheric composition stem from its advantages in reflecting global behavior. The most obvious advantage is the remoteness of major anthropogenic and natural terrestrial sources of carbon so that a higher degree of integration of surface exchanges is anticipated. Coupled with this advantage is the fact that the best avenues for reconstruction of past global behavior appear to involve the extraction of archived air from Antarctic ice.

AMS measurements of  $^{14}\text{C}$  have a potential role in elucidating mechanisms for temporal  $\text{CO}_2$  and  $\delta^{13}\text{C}$  variations observed in the ice core records, as well as in recent southern hemisphere spatial gradients for both species.

The application of AMS  $^{14}\text{C}$  measurements in ice cores has been spectacularly successful in quantifying the modifications to the concentrations and isotopic ratios of  $\text{CO}_2$  due to the diffusion and bubble formation processes in ice. The resulting 1000-yr atmospheric histories reveal subtle variations in global atmospheric  $\text{CO}_2$  of possible "natural" origins (the most recent example of which may have occurred through the early 1990s, and the most marked during the 1550–1800 AD period).

Interannual variation in spatial gradients in the isotopic composition of  $\text{CO}_2$  over the southern oceans is creating considerable difficulties in inversion studies aimed at deriving surface fluxes from atmospheric concentration measurements. Multi-species measurements, including AMS  $^{14}\text{C}$ , have the potential to distinguish between atmospheric and oceanic transport as causes for the variations.

## **SAMPLE PRESENTATION IN $^{14}\text{C}$ -AMS FOR BIOMEDICAL RESEARCH**

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$^{14}\text{C}$  samples for biomedical and environmental research continue to be measured at rates up to 300 per day on the one day per week that the CAMS spectrometer is available to us. The high demand and low spectrometer availability encourage the use of high-output graphite as our sample presentation method. The sealed-tube reduction of  $\text{CO}_2$  over zinc and titanium hydride on a cobalt catalyst is a reliable way to prevent sample-to-sample contamination and to retain the possibly high  $^{14}\text{C}$  concentration in a sealed or low-vapor-pressure form to avoid contaminations in the spectrometer build-

ing. We report on further performance parameters of this presentation mode, including a variable fraction of very high  $^{14}\text{C}$  concentration that occasionally appears at the start of many measurements. In pursuit of mating biochemical separation instruments to AMS, we have studied memory effects in a GC-type carbon analyzer and in a room-temperature  $\text{CO}_2$  adsorption material. Procedures are described for biological materials that we have found difficult to handle, such as urines with highly variable carbon contents.

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### **TRITIUM DEPTH PROFILING IN CARBON BY ACCELERATOR MASS SPECTROMETRY**

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For the measurement and depth profiling of hydrogen isotopes in the surface layers of solids, several methods have been developed. In Rossendorf, two methods have been successfully applied, elastic recoil detection analysis (ERDA) and nuclear reaction analysis (NRA). For the detection of very small amounts of tritium in carbon, such as deposited on the vessel walls of DD fusion experiments, the sensitivities of these techniques are not sufficient. A very sensitive method is accelerator mass spectrometry (AMS). A test facility for tritium detection by AMS was built at the Rossendorf 3 MV Tandatron. The Cs sputter ion source model 860-C is applied for generation of negative ions by sputtering the carbon tiles to be investigated. The negative tritium ions are selected with the injector magnet and are accelerated with the Tandatron at a terminal voltage of 1.5 MV. After acceleration and analysis with the switching magnet, the tritium ions are counted with a surface barrier detector, while the ion currents of deuterium and carbon are measured with a Faraday cup at the accelerator entrance. The residual HD and heavier ions are stopped in an Al foil in front of the detector. For well-defined depth profiling, the sample to be sputtered is screened by the holder with a 1.5-mm diameter opening and is mechanically scanned over a 3.0-mm diameter area by two off-axis disks rotating at different speeds. Due to this scanning the bell-shaped sputter crater is changed into a more cylindrical shape. After 3 h sputter time a crater depth of *ca.*  $20\ \mu\text{m}$  was measured. The tritium and deuterium depth profiles measured at a carbon tile from the diverter of the fusion experiment ASDEX-upgrade at the Max Planck Institut für Plasmaphysik in Garching are shown. The absolute isotope ratios T/H, including the isotope fractionation, are obtained by measurement of a standard sample from the Center for AMS of the Lawrence Livermore National Laboratory. The range of the isotope ratios T/H or T/D, which can be measured at our facility, is at present *ca.*  $10^{-6}$  to  $10^{13}$ . It is limited by the maximum counting rate of the detector and by the tritium contamination of the ion source, respectively.

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## THE DEVELOPMENT OF A GAS $\Delta E$ -POSITION SENSITIVE E DETECTOR FOR AMS STUDIES

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An AMS system at the University of Tsukuba has been realized by a molecular pilot beam method. A di- or tri-molecular beam is generated together with a particles of interest for the terminal potential control (Nagashima *et al.* 1993, 1994). This method essentially requires changing the charge state of the particles by passing through a thin foil in order to separate the pilot particles from the particles of interest. After passing through the foil, the particles diverge by multiple-scattering in the foil and consequently the size of a particle spot on a detector becomes large. In the case of  $^{36}\text{Cl}$  measurement, the diameter of a Cl spot is observed to be *ca.* 15 mm. Adding to this enlargement, the spot moves around the center by the influence of long-term instability of electronic instruments. Maximum fluctuation width observed is 8 mm. So, our AMS studies, at least, require a  $\Delta E$ -E detector with an entrance window of 23 mm in diameter. On the other hand, the large acceptance of the detector may cause unforeseen background increment. The background increment might be suppressed by using position information if these background particles are focused to a different position on the plane of the entrance window.

Here, a detector that consists of a gas  $\Delta E$  and a two-dimensional position-sensitive E counter has been developed. Diameter of the entrance window is 30 mm. A Capton window foil,  $7.5\ \mu\text{m}$  thick, is strong enough to keep  $\sim 40$  Torr ionization gas pressure without any foil supporter. The  $48 \times 48\ \text{mm}^2$  active area of the two-dimensional position sensitive E counter is big enough to cover the size of the entrance window and the particle divergence in the ionization gases. The detector is operated with iso-butane gasses. Pressure is precisely controlled by a gas flow system.

We will report the result of performance test, with  $\alpha$  sources as well as some preliminary results of radio-isotope detection.

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## AUTOMATED SYSTEMS AND TECHNIQUES UTILIZED AT THE NOSAMS SAMPLE PREPARATION LABORATORY: AN UPDATE OF PRODUCTIVITY AND QUALITY ISSUES

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The primary charge of the National Ocean Sciences AMS (NOSAMS) Facility at the Woods Hole Oceanographic Institution is to supply high-throughput, high-precision AMS  $^{14}\text{C}$  analyses for sea waters collected as part of the World Ocean Circulation Experiment (WOCE). To accomplish this we have automated as many sample preparation systems as possible. At AMS-6, preliminary laboratory automation was described (Cohen 1993), as well as external and internal checks on quality control issues pertaining to sample preparation (Osborne 1993). In July of 1995, automation of the graphitization system in the Sample Preparation Lab (SPL) was completed. Major improvements included a second  $\text{CO}_2$  splitting line (aliquots for  $^{13}\text{C}$ ,  $^{14}\text{C}$ , and archiving), a linear track system

(allows the automated cryogenic transfer of CO<sub>2</sub> from splitting line to graphite reactor), and automated data transfer to the SYBASE database upon graphite reaction completion (minimizes manual data entry). Through these developments we can now conduct up to 26 graphite reactions per day.

The SPL goal for 1996 is to become more productive through increased automated batch processing. In order to achieve this, we are presently duplicating the automated sea water stripping system to allow a two-fold increase in production (from 10 to 20 samples/day). Recent  $\delta^{13}\text{C}$  analyses of local sea water (internal standard) stripped of CO<sub>2</sub> in the SPL indicate the overall precision of the analysis is between 0.04–0.06‰. Work has begun towards automating the organic carbon combustion process, and automated batch processing of CaCO<sub>3</sub> hydrolysis reactions has recently been implemented. These systems will be described, as well as issues pertaining to insuring high quality results while increasing productivity.

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#### <sup>60</sup>Fe MEASUREMENTS WITH AN EN TANDEM ACCELERATOR

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The measurement of the long-lived radionuclide <sup>60</sup>Fe ( $\beta$ -decay, no  $\gamma$ ,  $t_{1/2} = 1.5 \times 10^5$  yr) is of interest for different applications. The main production mechanisms of <sup>60</sup>Fe are double neutron capture in <sup>58</sup>Fe and spallation reactions of copper or nickel. In astrophysics these processes are of importance in providing information (*e.g.*, from meteorites) about the early solar system and supernovae. The long half-life may also have implications on the management of waste produced by accelerators or nuclear reactors. The very low abundances of <sup>60</sup>Fe can only be measured with accelerator mass spectrometry. First experiments were made by Kutschera *et al.* (1984, 1986) with a FN tandem linac using a 360 MeV ion beam. Sample material from a beam dump (spallation of copper) and a meteorite were examined giving <sup>60</sup>Fe/Fe ratios of  $10^{-7}$  (Kutschera *et al.* 1984), resp.  $10^{-14}$  (Kutschera *et al.* 1986). In combination with a specific activity measurement, the first experiment was used for the determination of the half-life (Kutschera *et al.* 1984).

A new attempt to measure <sup>60</sup>Fe at low energies is being made using our EN tandem accelerator. At 6 MV the maximum yield from a carbon stripper foil in the terminal is for charge state 9<sup>+</sup>, resulting in an energy of 60 MeV. A pure iron sample (99.99%) gave unexpectedly high nickel rates (<sup>60</sup>Ni/Fe  $\approx 0.3$  ‰) in the detector. This can be explained by the much higher negative ion yield for nickel relative to iron and by contamination from ion source surfaces. Due to <sup>60</sup>Fe having a two units lower nuclear charge than stable <sup>60</sup>Ni, an isobar suppression in our gas ionization chamber of almost six orders of magnitude was possible. To reduce the high counting rates of nickel, we plan to use our

gas-filled magnet to separate the isobars spatially. A gain of another two orders of magnitude in isobar suppression is expected. The resulting background level will enable first measurements on samples from nuclear reactor material or from beam dumps of high energy accelerators.

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#### A COMPACT MULTI-SAMPLE HIGH-INTENSITY NEGATIVE ION SOURCE

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A high-intensity negative ion source based on the design of Middleton (Middleton 1984) and a compact 28-position sample changer were built and tested. The main modifications brought to the original source design are the following: 1) a Cs oven located inside the source body feeds Cs vapor to the ionizer shield region through a short (6cm) capillary tube with a high temperature gradient; 2) an external insulator, shielded by a cooled trap from Cs vapors, supports the cathode and sample changer assembly. The copper cathode holding the sample is cantilevered from its insulator, locked into precise position and cooled by freon circulation; 3) a 0.2-mm-thick Ta aperture shaped as an immersion lens for the Cs beam and thermally insulated from the cathode, was designed after Koslowsky *et al.* (1995) to reduce memory effects; and 4) the extractor insulator is protected from vapors by a concentric baffle. The fully pneumatic sample changer is composed of a rotary-actuated target wheel from which a cathode is pushed, locked into position by an actuator rod and retracted back into the wheel by taper friction. The wheel is directly indexed on its periphery using a precision Geneva-drive mechanism, eliminating any mechanical link with the rotary mechanism and allowing for fast drop-in loading. A gate valve insulates the vacuum in the source body during wheel loading. A viewing port allows for visual inspection of the samples and cathodes in the wheel. Negative ion beam currents obtained match or exceed ion sources of similar designs; current tests and experiments performed with the ion source will be reported.

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## NEW DETECTOR CONCEPTS FOR AMS

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We propose two new detector concepts that may be particularly well suited for AMS measurements with small accelerators: 1) a fully solid state  $\Delta E$ -E detector for light ions utilizing ion-induced electron emission from foils, and 2) a time-of-flight (TOF) system based on a two-dimensional electronic chopper.

### The Foil Detector

When a fast ion passes through a thin self-supporting foil, the yield of ion-induced electrons is a function of nuclear charge and velocity of the projectile, and is roughly proportional to the electronic stopping power  $dE/dx$ . Thus, particle identification is possible by measuring the number of electrons emitted with a multichannel plate and the projectile energy with a surface barrier detector. The use of a large number of foils (typically 10) greatly increases the detector efficiency and the resolving power.

### The Electronic-Chopper TOF System

In a typical TOF measurement in AMS, the start detector is a multichannel plate measuring ion-induced electron emission from a thin foil. In the present system, a two-dimensional electronic chopper is used. This avoids limitations due to energy straggling and charge-state changes in a foil. However, one has to find other means to generate the start signal in a well-defined relation to the ion's position. By applying harmonic voltages  $V_0 \sin \omega t$  and  $V_0 \cos \omega t$  to x- and y-deflection plates, respectively, a monoenergetic ion beam describes a circle on the detector located at some distance from the deflection plates. Let the TOF measurement be started at  $t = 0$ , and let an ion enter the deflection plates at an arbitrary time  $t = t_1$ . It then hits the detector after its TOF at some point with polar coordinates  $(r, j)$  on the circle, where it generates the stop signal. The radius  $r$  depends on the ion energy, the angular position  $j$  depends on  $t_1$ ,  $j = \omega t_1$ . To obtain the ion's TOF,  $t_1$  has to be subtracted from the measured time difference and hence,  $j$  has to be known. The experimental design can be greatly simplified, if the  $j$ -position is detected by a delay-line that is matched to the chopper period  $t = 1/\omega$  (delay time  $t_d = 0$  for  $j = 0$ ,  $t_d = t$  for  $j = 2\pi$ ). Thus, the measured time difference gives the ion's TOF without further correction. The particular advantages of this new spectrometer are very high duty cycle due to the continuous operation, and relatively low power requirements due to the harmonic excitation.

## THE NEW HVEE MULTI-ELEMENT AMS-SYSTEM

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During the last two decades, AMS has become an important tool in mass spectrometry for long-lived radioisotope measurements. Besides  $^{14}\text{C}$ , other elements such as  $^{129}\text{I}$  are of growing interest for research fields where the higher sensitivity of the AMS method is used instead of conventional counting techniques. Nowadays isotopes like  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$  and  $^{129}\text{I}$  can be measured with "small"

AMS-machines (terminal voltage  $\sim 3$  MV) down to the  $10^{-15}$  level with precisions of 5% down to 0.3%. Low-background and high-precision measurement requires specific AMS-techniques for each element. In order to design a multi-element system we have combined the advantages of the high-precision  $^{14}\text{C}$ -AMS spectrometer based on simultaneous injection (M.-J. Nadeau, this conference) and the flexible sequential injection for other elements. In this article the layout of the system and its physical characteristics will be presented.

## A PROBLEMATIC SOURCE OF ORGANIC CONTAMINATION OF LINEN

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Microscopic observations of a putative sample of linen from the Turin Shroud reveal that individual fibers of the cloth are surrounded by a bioplastic coating produced by bacteria-like microbes. The latter are thought to obtain their sustenance from carbon dioxide in the air. Fungal material is also found among the threads (Garza-Valdes *et al.* 1995). Similar observations have been made on various linen mummy wrappings. In particular, samples of the linen wrapping of an Egyptian mummified ibis dating between *ca.* 330 and 30 BC have been examined and the presence of this bioplastic coating was established. This coating is not removed in the standard acid-base-acid cleaning procedure that is recommended for cleaning cloth prior to carbon dating. The question is whether the presence of this coating materially affects the age of the cloth as obtained by carbon dating from what would be obtained if the actual cellulose of the flax from which the linen was made was dated. To obtain some answers to this question AMS radiocarbon measurements will be made at the NSF Arizona AMS Facility at the University of Arizona on the collagen in bone samples from this ibis and of samples of the linen wrapping of the ibis cleaned in the recommended fashion. In addition, it will be possible to obtain bone and linen wrapping samples from several human mummies owned by the Manchester Museum on which similar AMS  $^{14}\text{C}$  measurements can be made. If the measurements show that linen mummy wrappings are substantially younger than the associated bone collagen, procedures will be explored to recover glucose as a degradation product of the linen's cellulose. The ages of this cellulose and the associated bone collagen should be in agreement, barring the presence of cellulose-producing bacteria. It is worth noting that, because these bacterial infestations form surficial bioplastic coatings, they are most likely to have their maximum impact on the  $^{14}\text{C}$  measurements of the ages of cloth as opposed, for example, of parchment. The surface-to-volume ratio of cloth vastly exceeds that of other organic artifacts.

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## A NEW ACCELERATOR MASS SPECTROMETER FOR TRACE ELEMENT ANALYSIS AT THE NAVAL RESEARCH LABORATORY

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A new accelerator mass spectrometer (AMS) facility is under construction at the Naval Research Laboratory (NRL) to improve capabilities for trace element analysis of electronic and geologic materials. The trace element AMS (TEAMS) facility is based on an existing National Electrostatic Corporation 3-MV Pelletron tandem accelerator at NRL. The design goal is to provide parallel mass analysis over a broad mass range for conducting and insulating samples, 10- $\mu\text{m}$  lateral image resolution, depth profiling, and sensitivity to tens of ppt of trace impurities. To attain these goals, our TEAMS facility has incorporated a number of unique features.

We will use a modified commercial secondary ion mass spectrometer (SIMS) (Physical Electronics, model 6300) as the source of secondary ions. This instrument inherently provides our required imaging, depth profiling, and sample manipulation capabilities. The broad mass range of secondary ions generated by the SIMS source will be accelerated to 40 keV, then injected into the accelerator through a tunable Pretzel magnet notch-mass-filter (Knies, this conference). This unique magnet will simultaneously transmit from 1 to 200 amu of 40-keV ions by first dispersing the masses along the mid plane of the magnet, then recombining them as they exit. A flag system located along the mid plane will permit attenuation of intense matrix-related beams. The recombined ions will be focused to the object position of the accelerator with an Einzel lens.

We minimize background signals by providing low pressure ( $\sim 10^{-8}$  Torr) in the beam line and components, and by designing the ion optics to avoid small angle scattering of the beam. After acceleration, a single charge state of ions will be focused and selected by the combination of an electrostatic quadrupole triplet (EQT) and a  $3^\circ$  electrostatic bend. Only the selected ions will enter a large gap (41 mm) spherical electrostatic analyzer (ESA). An entrance aperture to the ESA will prevent the beam from striking its deflection plates. The 2.2-m-radius ESA will energy-analyze the beam through its  $30^\circ$  bend ( $E/\Delta E = 800$ ) and, combined with another EQT, focus the beam to the object position of an Enge split-pole magnetic spectrograph.

This spectrograph will provide parallel analysis over a broad mass range ( $M_{\text{max}}/M_{\text{min}} = 8$ ) with high resolution ( $M/\Delta M = 2500$ ). The spectrograph is double focusing with second-order-correct focusing along the 1.5-m-long dispersion plane. We expect a beam size of *ca.* 100  $\mu\text{m}$ . Initially, we will use only a few surface barrier detectors, but in the near future hope to obtain Si micro strip detectors to cover the entire focal plane. With appropriate high speed electronics, we will be able to produce multielement images of a sample with 10- $\mu\text{m}$  lateral resolution and 1-nm depth resolution. At this fine scale, sensitivity will be volume limited.

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**$^{10}\text{Be}$  INVESTIGATIONS OF SEDIMENTS, SOILS AND LOESS AT GNS***I. J. GRAHAM, R. G. DITCHBURN, R. J. SPARKS and N. E. WHITEHEAD*

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The past three years have seen rapid advancement in  $^{10}\text{Be}$  analytical methods at GNS, and the initiation of three research projects involving “garden variety”  $^{10}\text{Be}$  in surficial deposits. A study of nearshore marine sediments from the Wanganui Basin (Castlecliff Section) has identified a 6–8 fold increase in  $^{10}\text{Be}$  immediately above the location of the Brunhes-Matuyama paleomagnetic reversal, *ca.* 780 ka. A second  $^{10}\text{Be}$  “anomaly” of similar magnitude further up the sequence, *ca.* 700 ka, shows the increase in  $^{10}\text{Be}$  cannot be attributed solely to the paleomagnetic reversal, but in large part reflects a major global climatic change at those times resulting in an influx of  $^{10}\text{Be}$  during post-glacial ice melting.

The  $^{10}\text{Be}$  inventory of a soil formed on till on the inland edge of the Transantarctic Mountains (Antarctica) yields an apparent age of 80 ka (using average global  $^{10}\text{Be}$  flux). This age is very much less than the age of the surface estimated by geological and pedological reasoning, the difference being due to much lower rates of infall of  $^{10}\text{Be}$  onto the land surface than the global average value. An alternative interpretation of the data assuming constant  $^{10}\text{Be}$  flux and known decay rates suggests an age of 12–20 Ma, consistent with the geological evidence.

A 0–500 ka loess sequence at Rangitautau East (Wanganui) has been analyzed for  $^{10}\text{Be}$  content (both 4–53-m and <4-m sized separates). Interim results contrast with those of the Chinese Loess Plateau and hint at differences in  $^{10}\text{Be}$  flux rates between the Northern and Southern Hemispheres. The results are difficult to interpret in the absence of additional geochemical data, but show a broadly decreasing trend in  $^{10}\text{Be}$  (and  $^{10}\text{Be}/^9\text{Be}$ ) with increasing depth. The overall level of  $^{10}\text{Be}$  in these loesses is about twice that of loess from the Chinese Loess Plateau, reflecting a lower sedimentation rate and/or a higher  $^{10}\text{Be}$  source.

**THE LEIBNIZ-LABOR AMS FACILITY AT THE CHRISTIAN-ALBRECHTS-UNIVERSITÄT, KIEL, GERMANY***P. M. GROOTES,<sup>1</sup> H. ERLLENKEUSER,<sup>1</sup> A. GOTTDANG,<sup>2</sup> D. J. W. MOUSE,<sup>2</sup> M.-J. NADEAU,<sup>1</sup> J. M. SARNTHEIN,<sup>3</sup> M. SCHLEICHER<sup>1</sup> and H. WILLKOMM<sup>1</sup>*

The Leibniz-Labor für Altersbestimmung und Isotopenforschung of the Christian-Albrechts University accepted its new AMS facility from High Voltage Engineering Europa in September 1995. The system is based on a 3MV Tandatron with a single Cs ion source and a beam separator-recombinator for simultaneous injection of the three isotopic carbon beams similar to the NOSAMS Facility at Woods Hole Oceanographic Institution, USA, and the AMS system at the Centrum voor Isotopen Onderzoek at the University of Groningen, the Netherlands. The AMS system, its technical installations, and workshops are housed in a separate hall, adjacent to a building housing AMS sample- and target preparation laboratories and offices as well as the Kieler radiocarbon dating and mass spectrometry laboratories that are now part of the Leibniz Labor. We will present some of the specifications of the AMS system and its technical installations and discuss the new design features of the Leibniz AMS-machine, *e.g.*, the successful reduction of X-rays by permanent magnets. Examples of the system performance will be given. This includes results of the acceptance tests showing

that Poisson counting statistics at 0.15% and 0.22%, respectively, determined the statistical uncertainty in the  $^{14}\text{C}/^{12}\text{C}$  ratios measured for the individual samples of two test series.

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## DATING AND CORRELATING PROXY RECORDS OF CLIMATE CHANGE

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Evidence is increasing that significant climate fluctuations are essentially global in nature, though they may be recorded in ice cores, on land, and in deep-sea sediments with various delays and attenuation. Accurate, independent dating is therefore important to prove the synchronicity and correlation of events in different records of climate change. Once correlations have been established, the detailed layer-counts of the ice-core records from the summit of the Greenland ice sheet (back to ca. 50,000 yr) allow a refinement of the timing of events in other records, provided such events can be unambiguously correlated with the ice-core record.

I will discuss some of the new isotope results from Greenland and Antarctica and plausible ice-ocean-land correlations for the last glacial/interglacial cycle.

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## HIGH-RESOLUTION RADIOCARBON DATING OF A KODIAK ISLAND (ALASKA) LATE GLACIAL COLD EVENT

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A cold reversal at the end of the last deglaciation has been found at many sites around the globe. In most cases radiocarbon dates place those Younger Dryas type events between 11 and 10 kyr BP. An interesting question which we can address here is—were those events simultaneous to the Younger Dryas in Europe?

In the sediments recovered from lakes of Kodiak Island (Alaska) Peteet and Mann (1994) have found a cold and dry event which correlates with the Younger Dryas known from the North Atlantic region. Using high-resolution  $^{14}\text{C}$  dating of macrofossils selected from sediment of Phalarope Pond, we try to reconstruct changes in atmospheric  $^{14}\text{C}$  content that are known to have taken place during the Younger Dryas. A dramatic rise in atmospheric  $^{14}\text{C}$  content at the beginning of the Younger Dryas or  $^{14}\text{C}$  plateau (decreases in  $^{14}\text{C}$  content) at 10 kyr BP provides an opportunity for close correlation between paleo records of Kodiak Island and the European lakes.

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**ASSESSMENT OF THE AAA PRETREATMENT TECHNIQUE FOR CHARCOAL AND OTHER ORGANIC MATERIALS USED FOR <sup>14</sup>C AMS STUDIES***M. J. HEAD*

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Combustion of wood involves a complex series of physical transformations and chemical reactions that are further complicated by the heterogeneity of the substrate. Wood, and cellulosic materials in general, do not burn directly, but under the influence of sufficiently strong heat sources they decompose to a mixture of volatiles, tarry compositions, and highly reactive carbonaceous char. Gas-phase oxidation of the combustible volatiles and tarry products produces flaming combustion. Solid phase oxidation of the remaining char produces glowing or smouldering combustion, depending on the rate of oxidation.

The production of volatiles leaves a solid residue that is neither intact substrate nor pure carbon, but a different material at various stages of charring and carbonization. The intermediate chars are characterized by the functional groups present (including aromatic and olefinic structures); a high concentration of free radicals trapped in a rigid structure or stabilized by aromatic and olefinic structures; a large surface area; and a high degree of reactivity. All of these depend on progression of the secondary reaction in the solid phase.

Hence, wood charcoal and other organic components of sediments most likely contain relatively stable, but potentially active functional groups that may react with the surrounding micro environment relatively rapidly, or may lie dormant for a long period of time. Treatment of these types of materials with alkali in the presence of air could quite feasibly provide a mechanism for the incorporation of atmospheric CO<sub>2</sub> into the organic structure of the material to form carboxylate functional groups which would not be removed by further acid treatment. This treatment thus has the potential to incorporate younger carbon into the organic samples.

A series of charcoal samples have been analyzed for possible carboxylate functional groups using Fourier transform infra red spectrometry. These charcoals have then been given an acid-alkali-acid treatment without taking the precaution of providing an inert atmosphere, and the resulting residue has also been analyzed for possible carboxylate functional groups. A group of these samples (having apparent <sup>14</sup>C ages >20 k BP) both before and after treatment have been dated using <sup>14</sup>C AMS in order to gauge the possible contamination effect, and the results are presented. The potential for decarboxylation of these materials will be discussed.

**THE PROPERTIES OF HUMUS MATERIAL SEPARATED FROM PALAEOOLS IN LOESS-PALAEOLOS SEQUENCES FROM THE LOESS PLATEAU IN CHINA***M. J. HEAD,<sup>1</sup> W. J. ZHOU,<sup>2</sup> G. JACOBSEN,<sup>3</sup> and C. TUNIZ<sup>3</sup>*

The Loess Plateau of northern China contains loess-palaeosol sequences up to 200 m thick, formed over the last 2.5 myr. These sequences provide an extremely useful record of palaeoclimatic change within this area, and they have been quite well correlated with palaeoclimatic data from δ<sup>18</sup>O deter-

minations on foraminifera from deep-sea cores, and ice-core data from Greenland. However, it is extremely difficult to establish accurate chronologies for these sequences covering the last 30,000 yr.

These loess-palaeosol sequences are quite porous and are perched well above present-day water tables. The land surface is also subject to extensive agriculture. Consequently, the palaeosols (presently containing between 1 and 2% carbon) experience vadose conditions, with extensive amounts of water-soluble organic material passing through them to the water table below. The palaeosol matrix essentially consists of fine-grained reworked loess, with a relatively high clay content.

$^{14}\text{C}$  age determinations on the bulk organic material from palaeosols have been unreliable because of the incorporation of younger material. The separation of humic acids using alkali leaching techniques has provided material that seems to produce representative  $^{14}\text{C}$  ages. The molecular weights of these extracts vary between 1200 and 1800. A long-term study has been carried out on the chemistry of these "humic" extracts, and purification techniques have been developed to remove any low molecular weight organic components that have been adsorbed within the humic polymer structure. Solid state  $^{13}\text{C}$  NMR data indicate that these compounds basically consist of aromatic and carboxyl carbons, similar to those found in highly oxidized volcanic soils.

Further studies involving the use of UV radiation, ESR and FTIR spectra to determine the presence of stable free radicals have been carried out in order to determine the mechanism of formation of these humic "remnants", and to assess the viability of the purification techniques used.

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## AMS $^{14}\text{C}$ DATING OF LIME MORTAR

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There has long been great interest in possibility of radiocarbon analysis directly on mortar to date the construction time of buildings where no other type of material is available for dating or can be safely associated with the time of construction. Mortar has great potential as dating material, not only because of its ample availability and clear association with the construction to be dated but also because the carbonate formed from the burnt limestone (CaO) in the hardening process represents a sampling of the contemporaneous atmospheric  $\text{CO}_2$  content.

However, mortar is a very complex material, consisting of not only mortar carbonate but also unburnt fossil limestone inclusions and other carbon-bearing minerals and organic components. Attempts to apply conventional  $^{14}\text{C}$  dating has thus proven very difficult. Likewise, the seemingly obvious solution of utilizing AMS to selectively date the organic fraction has often led to exceedingly high ages (Tubbs and Kinder 1990). Strydonck *et al.* (1992) used both carbonate and organic fractions and found very scattered results.

Because of the urgent need for reliable mortar dating, we have continued work to develop a technique to isolate the pure mortar carbonate fraction from the rest of the crude mortar components. The preparation process includes not only mechanical and chemical separation but also mineralogical microscopy control of the success of this separation. So far, a large series (>100 samples) of dat-

ing results on different chemical fractions from a number of different (medieval) buildings have shown promising internal consistency. Some of these have been reported in Heinemeier *et al.* (1995).

The separation techniques and examples of mineralogical analysis will be presented along with series of dating results on medieval churches from Åland, Finland and, as a curiosity, the Newport Tower, Rhode Island, USA.

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## THE AMS FACILITY AT THE UNIVERSITY OF AARHUS, DENMARK

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The accelerator mass spectrometry (AMS) system based on the 6 MV EN tandem accelerator at the Institute of Physics, Aarhus is described. The current sample preparation methods, measurement procedures and system capacity for <sup>14</sup>C measurements are discussed. Information will be given on precision, accuracy and background level for different sample sizes and preparation techniques.

## IN-SITU PRODUCTION OF COSMOGENIC NUCLIDES: ACCELERATOR SIMULATION EXPERIMENTS WITH MUONS AND MEASUREMENT OF DEPTH PROFILES

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The *in-situ* production of cosmogenic nuclides is important for the determination of background events in all low-level detection experiments (e.g., the experiment <sup>205</sup>Tl (ν<sub>e</sub>, e<sup>-</sup>) <sup>250</sup>Pb (Neumaier, Nolte and Morinaga 1991), for many geophysical applications (e.g., the determination of erosion rates or dating of old groundwaters) and for industrial applications. In the present paper, the *in-situ* production of cosmogenic radionuclides was investigated by performing accelerator simulation experiments with slow negative muons at PSI Villigen and with 200 GeV muons at CERN and by measuring concentrations of <sup>10</sup>Be and <sup>26</sup>Al in natural quartz samples up to depths of 260 m.

The *in-situ* production rate was calculated as a function of depth z taking into account spallation reactions, reactions with stopped negative muons, reactions with fast muons and background reactions. The production rate due to μ<sup>-</sup> capture can be expressed by  $P_{\mu^-}(z) = I_{\mu^-}(z) \cdot f_C \cdot f_D \cdot f^*$  with the

rate of stopped negative muons  $I_{\mu^-}(z)$ , the chemical compound factor  $f_C$ , the probability of nuclear capture  $f_D$  and the probability  $f^*$  of the reaction channel after nuclear  $\mu^-$  capture to the investigated nuclide. The production due to fast muons is given by  $P_{\mu^{\text{fast}}}(z) = \Phi_{\mu^{\text{fast}}} \cdot \sigma$  with the flux of fast muons  $\Phi_{\mu^{\text{fast}}}$ . The energy-dependent cross section  $\sigma$  is given by  $\sigma = \sigma_0 \cdot E_{\text{mean}}^{0.7}$  according to the Wolfendale rule with the mean muon energy  $E_{\text{mean}}$  in GeV at the considered depth.

With the PSI irradiations, the following probabilities  $f^*$  of particle emission channels after  $\mu^-$  capture have been measured:  $f^* = (5.2 \pm 0.6) \cdot 10^{-3}$  for  $O(\mu^-, \nu_{\mu}, \alpha, \text{pxn})^{10}\text{Be}$  [ $x = 1-3$ ], and  $f^* = (1.4 \pm 0.4) \cdot 10^{-3}$  for  $S(\mu^-, \nu_{\mu}, \alpha, \text{xn})^{26}\text{Al}$  [ $x = 2-4, 6$ ]. From the result of the oxygen irradiation and the earlier result obtained for  $^{26}\text{Al}$  from the irradiation of quartz (Strack *et al.* 1994), the production ratio of  $^{26}\text{Al}$  to  $^{10}\text{Be}$  after  $\mu^-$  capture in quartz has been deduced to be  $P(^{26}\text{Al})/P(^{10}\text{Be}) = 7.3 \pm 1.5$  (Heisinger *et al.* in preparation). In another experiment, the channel probability from Ca to  $^{36}\text{Cl}$  was measured.

For the determination of the fast muon cross section  $\sigma_0$  of  $^{10}\text{Be}$  (from O),  $^{14}\text{C}$  (from O),  $^{26}\text{Al}$  (from Si, S and Al),  $^{36}\text{Cl}$  (from Ca),  $^{53}\text{Mn}$  (from Fe) and  $^{250}\text{Pb}$  (from Tl), several targets have been irradiated at CERN. This work is still in progress.

Depth profiles of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in natural quartz samples were measured in northern Bavaria up to depths of 260 m and were compared with calculated profiles taking into account erosion. From this comparison, the erosion rate in the last million years was determined to be *ca.*  $5 \mu\text{m yr}^{-1}$  (Heisinger *et al.* in preparation).

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## USING COSMOGENIC RADIONUCLIDE CONCENTRATIONS TO DETERMINE GLACIAL EROSION ACROSS ALPINE VALLEYS

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Recent work has shown the value of cosmogenic radionuclide techniques in addressing substantive questions in geomorphic processes and landscape histories. In addition to exposure ages, the  $^{36}\text{Cl}$  technique can be used to determine erosion rates and the extent of soil/snow/tree cover since initial exposure of a rock surface. Recent advances at PRIME Lab have focused on isolating the neutron-activation production component from the spallation component using mineral separates. This new technique provides data that can be used to determine both the erosion rate and exposure age of surface samples.

A field sampling program has been designed with the aim of using this new  $^{36}\text{Cl}$ - $^{36}\text{Cl}$  AMS method to yield insight into rates and patterns of glacial erosion. As part of this we have calculated distribu-

tions of bedrock-surface radionuclide concentrations across a glaciated valley as a function of glacial erosion rates and patterns. Using equations describing the build up of radionuclides with depth for a rock undergoing erosion, we can predict valley-scale cross-sectional patterns of radionuclide concentration at bedrock surfaces, for a range of hypothetical patterns and rates of glacial and non-glacial (beyond the ice margin) erosion. To validate our calculations we collected samples along a cross-section of a glacial valley in the Wind River Range, Wyoming. The collected bedrock samples are being analyzed by AMS for  $^{36}\text{Cl}$ . Results of our calculations suggest that cosmogenic radionuclide techniques have the potential to accurately constrain both relative and absolute erosion rates, as well as patterns of glacial and non-glacial erosion.

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### **THE $^{129}\text{I}$ MEASUREMENT PROGRAM AT THE ANTARES AMS CENTER**

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An  $^{129}\text{I}$  measurement capability has been developed at the ANTARES AMS Centre. Measurements have been performed using the  $11^+$  charge state at 6.5 MV with C foil stripping. The key components that minimize the interference from  $^{127}\text{I}$  in the detector are the high-resolution injection magnet ( $M/\Delta M \approx 200$ ) and the  $22^\circ$ , 5m radius electrostatic analyzer. The detection system incorporates a 600mm time-of-flight system (TOF) and a Si detector to measure total energy. We have achieved a TOF resolution of 500ps, which is sufficient to resolve the  $^{129}\text{I}$  from remaining  $^{127}\text{I}$  interference. A background corresponding to a  $^{129}\text{I}/^{127}\text{I}$  ratio of  $1.5 \times 10^{-13}$  has been achieved.

We have been funded by the Australian Safeguards Office to develop this capability and measure anthropogenic  $^{129}\text{I}$  as part of an international programme under the auspices of the IAEA. This has required the development of techniques to extract iodine from a variety of sample materials including water, sediments and biological materials. We are also currently engaged in a research project aimed at the measurement of naturally occurring  $^{129}\text{I}$  in iodoargyrite in an attempt to date the formation of the secondary zone of the orebody at Broken Hill, Australia.

### **DESIGN OF A COMPACT 1 MV AMS SYSTEM FOR BIOMEDICAL RESEARCH**

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The acceptance of accelerator mass spectrometry in biomedical research will require the development of cost-effective, laboratory-sized AMS systems which can be used in conjunction with conventional gas and liquid phase separation techniques. This paper describes the design of a low-energy AMS system optimized for the detection of  $^{14}\text{C}$  and  $^3\text{H}$  in organic samples. The system utilizes a compact 1 MV tandem accelerator that incorporates a foil stripper and a solid-state high-voltage switching power supply developed by Newton Scientific, Inc. (NSI). The low energy analyzer, accelerating column, and high-energy analyzer are designed for efficient transport and analysis of



both carbon and hydrogen beams using the minimum number of optical elements. The resulting instrument is very compact: the entire AMS system including the injector, ion source and high energy analyzer is just under 3 m wide and is *ca.* 1.3 m high and 1 m deep. The relatively small size of this system will allow its installation in most biomedical laboratory facilities.

The NSI AMS system is based on stripping of  $^{14}\text{C}$  to the  $2^+$  charge state at approximately 1 MeV using a thin carbon foil. We have obtained new experimental results demonstrating that foil stripping at this energy results in the elimination of molecular interferences in the  $2^+$  charge state to a level more than adequate for detecting  $^{14}\text{C}$  at or near contemporary levels. The use of a carbon foil stripper is advantageous because of its simplicity and short insertion length; however, it introduces greater scattering and energy spread compared with a gas stripper. These effects have been analyzed through experiments and computer simulations and the results used to determine the required acceptance of the beam optical components downstream of the stripper.

Performance of the NSI AMS system in conjunction with a gas chromatograph has been evaluated for the analysis of post-labeled organic samples of interest in toxicology research. The system is predicted to provide high measurement accuracy for the quantitation of sample sizes at the attomole level. Quantitation of sample sizes down to the zeptomole level will also be attainable, with the accuracy limited by the counting statistics on the small number of detected  $^{14}\text{C}$  atoms. The system design and performance analysis will be presented.

This work was supported by the National Institutes of Health under Grant No. R43CA66400 and the National Science Foundation under Grant No. DMI-9461636.

## **EXPERIMENTAL INVESTIGATION OF THE DESTRUCTION OF 1 MeV $^{12}\text{CH}_2$ MOLECULES IN SINGLE AND DOUBLE STRIPPING FOILS**

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Recent interest in the use of accelerator mass spectrometry for the analysis of biomedical samples has led to the design of compact AMS systems using tandem accelerators with terminal voltages of 1 MV and below. Detection of  $^{14}\text{C}$  in the  $2^+$  charge state is advantageous for a low-energy AMS system because of the increased transmission efficiency and reduced system complexity compared with detection of the  $3^+$  charge state using the same terminal voltage. We have performed experiments at the IsoTrace Laboratory examining the destruction of mass-14 interfering molecules in the  $2^+$  charge state after passage through both single and double carbon stripping foils at 1 MeV. For single foils with thicknesses of 2–3  $\mu\text{g}/\text{cm}^2$ , the molecular background ratio was measured to be <20% of contemporary  $^{14}\text{C}$  with no detector energy resolution. Use of a thin detector entrance foil to provide energy dispersion between the surviving molecules and  $^{14}\text{C}$  ions, coupled with threshold energy discrimination, was shown to reduce this background level to 5% of contemporary  $^{14}\text{C}$ , while allowing detection of 95% of the incident  $^{14}\text{C}$  ions.

It has been suggested that, if molecular survival can be attributed primarily to the wake-riding effect, then the use of a foil pair may provide much higher molecular attenuation than a single foil of equal thickness (K. H. Purser and A. E. Litherland, personal communication). The results of experiments

performed using pairs of carbon foils conclusively demonstrate, for the first time, that the passage of molecular ions ( $^{12}\text{CH}_2$  and  $^{13}\text{CH}$ ) through a pair of thin carbon stripping foils at 1 MeV results in significantly increased attenuation of molecular ions in the  $2^+$  charge state as compared to passage through a single carbon foil of comparable total thickness. The detected molecular counts after passage through a single  $4.4 \mu\text{g}/\text{cm}^2$  foil correspond to a surviving fraction of  $7\text{--}47 \times 10^{-11}$ , depending on the assumed energy and angular spread introduced by the foil. No statistically significant molecular survival was observed for pairs of stripping foils with a combined thickness of  $4.0\text{--}4.6 \mu\text{g}/\text{cm}^2$ . The upper limit on surviving molecular fraction for the double foils was a factor of 15 lower than for a single foil of comparable thickness. These results show that a low-energy AMS system based on the detection of  $^{14}\text{C}$  in the  $2^+$  charge state after a foil stripper will provide more than adequate discrimination for the analysis of biomedical samples labeled at or above contemporary  $^{14}\text{C}$  and that low-energy systems employing double foils may be potentially useful for radiocarbon dating.

This work was supported by the National Science Foundation under Grant No. DMI-9461636 and the National Institutes of Health under Grant No. R43CA66400.

### MEASUREMENTS OF PROTON- AND NEUTRON-INDUCED PRODUCTION CROSS SECTIONS FOR $^{36}\text{Cl}$ FROM Ca AND K

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Production cross sections for  $^{36}\text{Cl}$  have been measured for the nat.K(p,x),  $^{39}\text{K}$ (p,x), nat.Ca(p,x), and  $^{40}\text{Ca}$ (p,x) reactions. In addition to these proton reactions, neutron cross sections up to 40 MeV for the nat.K(n,x) and nat.Ca(n,x) reactions have also been measured.

Radionuclides produced in extraterrestrial surface materials by energetic solar flare particles provide clues about the history of solar activity. Changes in solar activity are reflected in the particle intensities and energy spectra averaged over the mean lives of the radionuclides measured (Reedy *et al.* 1983). Recently Nishiizumi *et al.* measured a  $^{36}\text{Cl}$  depth profile in lunar surface rock 74275 (1991) and 64455 (1995). The observed profiles of  $^{36}\text{Cl}$  in the top several  $\text{g}/\text{cm}^2$  were discussed with the constancy of solar energetic particle flux and spectrum using new  $^{36}\text{Cl}$  production cross sections. Since investigation of the solar flare energetic particle intensity over the past half million years absolutely requires  $^{36}\text{Cl}$  production cross sections from the major target elements Ca and K,  $\text{CaCO}_3$  and  $\text{KNO}_3$  targets were bombarded with both protons from the INS SF cyclotron and semi-monoenergetic neutrons (produced via the  $^7\text{Li}$ (p,n) reactions using the same cyclotron). For the proton bombardments we used both natural and isotopically enriched targets. These targets were subsequently chemically processed after adding Cl carriers, following which the  $^{36}\text{Cl}$  activities were measured by AMS at LLNL.

Figures 1 and 2 (not given here) show the proton-induced production cross sections for Ca and K targets, respectively. Figure 1 also displays the data recently reported by Shiekkel *et al.* (1994). We will present neutron-induced cross sections as well.

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## **FORAMINIFERAL <sup>14</sup>C RECORD OF THE YOUNGER DRYAS IN SANTA BARBARA BASIN, ODP HOLE 893A**

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Radiocarbon dating by accelerator mass spectrometry of coexisting planktonic and benthic foraminifera separated from Santa Barbara Basin sediments was used to provide a chronology for the upper 70 m of core from Ocean Drilling Program Hole 893A. The <sup>14</sup>C chronology allows a determination of the age of the Younger Dryas (YD) in Santa Barbara Basin, and provides evidence for changes in ocean circulation in the eastern Pacific over the past 20,000 yr. The YD event is represented in Santa Barbara Basin by a climatic change based on oxygen isotopic shifts measured in planktonic and benthic foraminifera, and occurs in a non-laminated interval within laminated sediments at a depth of 17.60 to 20.40 mbsf. The event was dated using six planktonic foraminifer samples. The calibrated <sup>14</sup>C age for the initiation of the YD in Santa Barbara Basin is 12,970 yr BP, coincident with that in North Atlantic deep-sea cores, glacial deposits in New Zealand, and ice-core records. The <sup>14</sup>C age for the termination of the YD in Santa Barbara Basin is poorly constrained due to a 1300-yr <sup>14</sup>C plateau, falling between 11,000 and 12,300 yr before present. Assuming a constant sedimentation rate in the core, the termination age would be 11,220 yr BP, with a duration of 1750 yr. During the YD, the planktonic-benthic age difference averaged 90 yr, compared with an average value of 470 yr in younger and older sediments. The age decrease of surface to bottom waters during the YD may reflect a change in source of Pacific Intermediate Water, with a greater proportion originating from a more proximal source.

## **CHEMICAL BEHAVIOR OF Be AND Al DURING AMS TARGET PREPARATION INVESTIGATED WITH CHEMICAL SPECIATION CALCULATIONS**

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The chemical extraction of Be and Al from quartz for surface exposure dating studies is not only time-consuming but very crucial with respect to the final exposure age and associated errors. The complex multi-stepped procedure is often treated in a black-box-like manner; it is not always clear in what chemical form the Be and Al exist. As a matter of fact, it is not difficult to "mysteriously lose" one or the other in the course of the extraction. Chemical speciation calculations reveal the chemical form and thus subsequent behavior of both Be and Al during each of the extraction steps.

Such chemical calculations can be used to predict the interferences resulting from an excess of, *e.g.*, fluoride or unwanted major cations (Mg, Ca, Fe), as well as the behavior of the different cations during ion exchange processes. As an example, in our experiences with some of the weakly metamorphosed granites from the Swiss Alps, inclusions and intergrowths of muscovite or epidote are very difficult to separate from the quartz. The presence of just a few of these inclusions can yield, *e.g.*, Mg and Ca in the milligram range, greatly affecting both the column efficiency and the conditions for precipitation. After a quick check of the chemical make-up of the sample solution with the ICP-AES, one can model the extraction steps with a chemical speciation program. In this way one can head off problems by adding additional extraction steps, thus tailoring the preparation uniquely to each sample.

#### **PROBLEMS IN THE CHRONOLOGY OF THE GLACIATIONS OF THE SWISS ALPS ADDRESSED WITH SURFACE EXPOSURE DATING**

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Questions about the exact timing of the Last Glacial Maximum (LGM) in Switzerland as well as on the synchronicity of the maxima of the largest glaciers, *e.g.*, the Rhone and the Rhine, remain unanswered. Huge erratics dot the Swiss foreland and mark the furthest extent of the respective glacial lobe. We have begun to exposure date some of these erratics. By using <sup>10</sup>Be, <sup>26</sup>Al and <sup>36</sup>Cl, limitations imposed by rock type are avoided.

The post-LGM glacial collapse was punctuated by a sequence of readvances which left behind moraines of, *e.g.*, the Gschnitz, Clavadel, Daun, and Egesen stands. As a first step we have dated blocks found along the crest of an Egesen moraine at Julier Pass. Egesen moraines are thought to be coupled with the Younger Dryas (11,000 to 10,000 <sup>14</sup>C yr ago), based on geomorphological considerations, equilibrium line depression values, regional correlation, and radiocarbon dating. The suite of six boulders we have dated yielded two distinct exposure ages for the double-walled moraine of the Lagrev glacier. The outer moraine has an exposure age of *ca.* 12,000 yr while the inner has an age of 10,300. Taken together the moraine complex has an exposure age of 11,000 yr.

The final stage of the demise of the glaciers was their disappearance from the high mountain passes. We are dating a series of samples from Grimsel Pass to pinpoint the time of its deglaciation. This provides more information about the timing of the warming-up and the rate of deglaciation at the end of the LGM and can be compared with pollen information on influx of pioneer vegetation into the mountain valleys.

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## ASPECTS OF TARGET PREPARATION AT THE ANTARES AMS CENTER

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To date, the ANTARES Target Preparation Laboratories have concentrated on setting up the procedures for processing carbon samples for AMS measurement. Graphite targets are prepared by the reduction of carbon dioxide over an Fe catalyst using Zn as the reducing agent (Slota *et al.* 1987). Samples containing 0.3 mg or more of original carbon are processed routinely for radiocarbon analysis. The current  $^{14}\text{C}$  chemistry background for 1 mg carbon is ~0.3% of modern carbon. With the increasing demand for measurement of extremely small samples, we are currently investigating methods for the preparation of targets containing tens of micrograms of carbon.

We are also expanding the capabilities of the laboratories to encompass the preparation of targets for the measurement of other isotopes such as  $^{129}\text{I}$ ,  $^{36}\text{Cl}$ ,  $^{10}\text{Be}$  and  $^{26}\text{Al}$ .

The difficulties associated with the handling and digestion of iodine samples is well documented. Foti (1979) has shown that losses can occur in the collection and drying of the sample and there have been numerous and conflicting recommendations for the ashing or digestion of samples for iodine analysis (Bock 1979; Muramatsu, Ohomo and Christoffers 1984; Marchetti, Rose and Strume 1994). At present this group is testing procedures for the extraction of iodine from a variety of environmental samples.

In addition, this laboratory is also setting up procedures for the extraction of *in-situ*  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$  from terrestrial rocks. A laboratory in a separate building is to be used for the preparation of  $^{14}\text{C}$  samples for a biomedical tracer experiment. A sealed-tube technique for combustion and graphitization is presently being tested, following Vogel (1992).

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## STUDY OF ALUMINUM MEDICAL APPLICATION BY $^{26}\text{Al}$ TRACER

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Pneumoconiosis remains the most serious occupational disease in China. Therefore, it is a very important research program to prevent and treat such disease in the field of occupational Health.

In the last several decades, a lot of research has been done on the prevention and treatment effects of aluminum citrate on pneumoconiosis, indicating that aluminum citrate is effective and with fewer

side effects. But, its mechanism has still been poorly understood so far. For this purpose, a collaborative program using  $^{26}\text{Al}$  as tracer is underway between the AMS Laboratory of the Chinese Institute of Atomic Energy and the Department of Occupational Health at Beijing Medical University.

The focus of this study is to explore the retention difference of Al in rat lungs, depending on whether it was administrated intratracheally singly or with  $\text{SiO}_2$ . The rats were sacrificed 2 or 7 days later following the administration and the amount of  $^{26}\text{Al}$  in the lungs and blood serum was determined.

#### DETERMINATION OF THE HALF-LIFE OF $^{79}\text{Se}$ WITH THE ACCELERATOR MASS SPECTROMETRY TECHNIQUE

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Half-lives of radioisotopes are important nuclear data and have applications in science and technology. Due to technical problems, half-lives of some of radioisotopes, in particular, long-lived radioisotopes, have had values with large uncertainty; the  $^{79}\text{Se}$  half-life is one of those. In the recent chart of the nuclides the half-life of  $^{79}\text{Se}$  has an upper limit of only  $6.5 \times 10^4$  a.

In this paper we present a measurement of the half-life of  $^{79}\text{Se}$ . The number of  $^{79}\text{Se}$  atoms was determined from the measured  $^{79}\text{Se}/\text{Se}$  ratio using the accelerator mass spectrometry (AMS) system at the China Institute of Atomic Energy, and the decay rate of  $^{79}\text{Se}$  in the samples was determined by counting the emitted  $\beta$  rays with the liquid scintillation spectrometer. The  $^{79}\text{Se}$  was separated from the high-level waste of a nuclear fuel reprocessing plant. The extracted Se ( $^{79}\text{Se}$ ) was transformed into CdSe for use in the AMS measurement of the  $^{79}\text{Se}/\text{Se}$  ratio. The main problems in the determination of  $^{79}\text{Se}$  with the AMS system are the isobaric and isotopic interferences. The interfering isotopes,  $^{78}\text{Se}$  and  $^{81}\text{Se}$ , can be separated using an electrostatic deflector. However, the isobar,  $^{79}\text{Br}$ , cannot be separated by the accelerator mass spectrometry system, due to energies (*ca.* 100 MeV) that are not high enough for the separation. Fortunately, bromine has two isotopes,  $^{79}\text{Br}$  and  $^{81}\text{Br}$ , with well-known abundance in natural materials, and  $^{81}\text{Br}$  is the only stable nuclide having a mass number of 81 in nature. We have deduced the  $^{79}\text{Br}$  interference from the  $^{81}\text{Br}$  measurement in the same samples. From the measured decay rates and number of atoms for  $^{79}\text{Se}$  in three samples, the half-life was deduced to be  $(1.1 \pm 0.2) \times 10^6$  a. Additionally, we have re-examined the half-life of  $^{79}\text{Se}$  reported in Parker *et al.* (1949) and the upper limit of the half-life was re-calculated to be  $6.5 \times 10^5$  a, not  $6.5 \times 10^4$  a as reported in the original paper.

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#### AMS IMAGING OF RADIOCARBON DISTRIBUTION IN RAT BRAIN

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Radiocarbon has been an important labeling element in biological metabolism studies. By interfacing an accelerator mass spectrometer (AMS) with a scanning microprobe secondary ion source, it is possible to image the  $^{14}\text{C}$  distribution in a biological sample at high resolution (1 micron), high sen-

sitivity and in a short time. The AMS at Oxford is normally dedicated to  $^{14}\text{C}$  measurements and detects secondary  $^{14}\text{C}^-$  ions as single  $^{14}\text{C}^{3+}$  ions with an overall efficiency of *ca.* 25–30%.

We studied biological samples from rat brain microinjected *in-vivo* with a  $^{14}\text{C}$ -labeled neurotransmitter GABA (4-amino-n-[U- $^{14}\text{C}$ ] butyric acid), which is selectively taken up by neurons having GABA carriers and using GABA as transmitter, and also by glial cells. The brain tissue was perfused and fixed with a mixture of paraformaldehyde and glutaraldehyde, in order to bind covalently the accumulated transmitter to proteins, then embedded in epoxy resin and sectioned serially at  $0.5\ \mu\text{m}$  using standard histological procedures. The two adjacent sections to those used for AMS imaging were either immunolabeled with antibodies to GABA to reveal GABA-containing cells, or stained with toluidine blue in order to visualize every cell, so that the distribution of  $^{14}\text{C}$  revealed by AMS could be matched to the distribution of all cells. The average concentration of radiocarbon ( $^{14}\text{C}/^{12}\text{C}$ ) was measured to be  $10^{-6}$ . By simultaneously measuring the  $^{14}\text{C}$ ,  $^{13}\text{C}$  and  $^{12}\text{C}$  signals, we were able to demonstrate that the localized peaks of radiocarbon activities could be readily identified and matched to GABA-immunopositive neurons and glial cells by aligning the radiocarbon-deficient blood vessels with the vessels in the adjacent histologically stained section. The results revealed the selective uptake of the neurotransmitter, GABA. The technique compares favorably with high-resolution autoradiography. Other applications in neuroscience at cellular and subcellular level using radiocarbon imaging by AMS will also be discussed.

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## MEASUREMENTS OF ALUMINUM TRANSPORT IN WHEAT AT THE CELLULAR LEVEL

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AMS can be used to address a longstanding and serious agricultural problem, namely, aluminum toxicity in a crop species, wheat. Understanding the toxicity mechanism will allow management strategies for dry-land soils laden with aluminum. Numerous mechanisms of Al toxicity and tolerance in plants have been proposed in recent years (Foy 1988; Roy *et al.* 1988; Mehrg 1991) but a clear consensus is not evident mostly because of a lack of a good measurement technique. Even the location of the primary lesion resulting from aluminum exposure has yet to be determined.  $^{26}\text{Al}$  is the only tracer isotope that can register the evolution of aluminum in plants. We have studied the transport of aluminum through the plasma membrane of wheat cells and its accumulation by tracing  $^{26}\text{Al}$  with AMS. We have observed residence times for the aluminum in several compartments and have noted distinct differences between aluminum-tolerant and aluminum-sensitive wheat cultivars Maringa and Katepwa, respectively. Young wheat plants are exposed for 24 hr to tracer quantities of  $^{26}\text{Al}$  (0.2 ng/l) introduced in a  $50\ \mu\text{M Al}_3^+$  plus nutrient bath. The plants are removed from the solution and placed in a fresh tracer-free nutrient solution. The solution, after elution from the plant roots, is collected and replaced at timed intervals varying from 10 sec to over 8 hr. A carrier solution of  $^{27}\text{Al}$  is added to the samples and  $\text{Al}_2\text{O}_3$  samples processed from the solutions are analyzed at the Rehovot Koffler 14UD Pelletron. The amount of aluminum released versus time is reflected in the measured  $^{26}\text{Al}/^{27}\text{Al}$  ratios that vary between *ca.*  $10^{-10}$  to  $10^{-13}$  in two very striking steps. There is a 100-fold drop in the aluminum release in the first 40 min followed by a very long time constant release over the following 8 hr. Both aluminum-sensitive and aluminum-tolerant varieties exhibit this behavior with the sensitive plants possessing much more residual aluminum in the long time

constant compartment. The utility of AMS to study the transport of aluminum in plants has been demonstrated. The sensitivity of the technique allows standard plant cell experimental manipulation techniques to be applied to agriculturally relevant cultivars. This allows a direct understanding of aluminum toxicity in agriculture at a cellular level.

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#### RADIOCARBON AMS DATING OF THE THERMOPHILOUS MOLLUSC SHELLS FROM PETER THE GREAT GULF COAST, RUSSIAN FAR EAST

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We have studied the <sup>14</sup>C age of thermophilous molluscs (*i.e.*, species that were not found alive in the area since the 1930s, but occur now *ca.* 500 km southward) in Peter the Great Gulf, northwestern Sea of Japan. The <sup>14</sup>C AMS dating of individual shells of three thermophiles, *Andara subcrenata* (Lischke), *A. inaequalvis* (Bruguière) and *Meretrix lusoria* (Röding), based on 1994 fieldwork around Vladivostok and the Posiet Bay. The shell samples were collected from both coastal outcrops and shell middens of the Early Neolithic Boisman culture. The *Yoldia johanni* individual shell collected alive in the Wrangel Inlet in 1932 (submitted for dating by K. A. Lutaenko 1994) gave the range 505 ± 30 BP (OS-3024). This value is the reservoir correction for the area and should be applied to all the ages shown here.

The results of <sup>14</sup>C dating may be subdivided into three groups in terms of age. The first and largest group contains <sup>14</sup>C dates corresponding to the Holocene Climatic Optimum. There are all the dates made on *Meretrix lusoria* shells: Vostok Bay (submitted by G. A. Evseev 1994), 6460 ± 50 BP (OS-3032); Boisman 1, 5690 ± 45 BP (OS-3030); Boisman 2, 6070 ± 35 BP (OS-3031), and 6140 ± 40 BP (OS-3033). The majority of both *Andara subcrenata* and *A. inaequalvis* samples also have similar ages: Talmi Lagoon core, 4760 ± 30 BP (OS 2319) and 4770 ± 30 BP (OS-2320); Talmi Lagoon section, 5320 ± 45 BP (OS-3020); Troitsa Inlet, 6280 ± 35 BP (OS-3029), and 7010 ± 55 BP (OS-3021), Boisman 2, 7640 ± 35 BP (OS-2318). The second group (shells of *A. inaequalvis*) corresponds to Subboreal and Early Subatlantic periods: Amur Bay, 2750 ± 35 BP (OS-3023); Ussuri Bay, 1760 ± 45 BP (OS-3022). The third group is represented by only one date (*A. inaequalvis*) and may be considered as modern (*ca.* 100 <sup>14</sup>C yr old due to reservoir correction of *ca.* 500 yr.): Amur Bay, 690 ± 25 BP (OS-3025).

The first results of <sup>14</sup>C AMS dating of thermophiles on Peter the Great Coast show that *A. inaequalvis* does not appear to be restricted to the mid-Holocene; rather, the dates obtained seem to suggest that it may still be alive in the area. Previously, the results of conventional <sup>14</sup>C dating gave only mid-Holocene dates (see review in Lutaenko 1993).



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## CHLORINE-36 IN AUSTRALIAN RAINFALL: HYDROLOGICAL AND ATMOSPHERIC IMPLICATIONS

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The applications of <sup>36</sup>Cl in hydrology require an understanding of its deposition on the Earth's surface. The atmospheric production of <sup>36</sup>Cl by cosmic-ray spallation on <sup>40</sup>Ar suggests that <sup>36</sup>Cl could also be useful as a tracer of such processes as stratosphere-troposphere exchange. We report on a survey of the <sup>36</sup>Cl composition of atmospheric precipitation from remote areas of Australia, in terms of their impact on the use of <sup>36</sup>Cl as a hydrological and atmospheric tracer.

Bulk precipitation samples were collected from two rainfall-collection arrays at three-monthly intervals over two years: the WE array (10 sites) extended in a west to east direction from the coast of Western Australia south of Geraldton, inland to Warburton in Central Australia, and the SN array (8 sites), extended in a south to north direction from Port Lincoln in South Australia to Kakadu in the Northern Territory.

With the exception of the tropical sites, which are anomalously high, the mean <sup>36</sup>Cl fallout in atoms/m<sup>2</sup>/s follows the form of latitude dependence predicted by Lal and Peters (1967), but at levels 1.4 × greater than predicted. This is in contrast to measurements in the northern hemisphere, which require an even larger normalization factor of 3–4. We believe the difference is due to enhanced stratosphere-troposphere exchange in the northern hemisphere, promoted by the greater area of landmass and the predominantly asymmetric thermal structure of the northern atmosphere.

At non-tropical latitudes, maximum fallouts were observed in spring, presumably due to the seasonal changes in the height of the tropopause, in agreement with northern hemisphere measurements.

The anomalously high fallout in northern Australia occurs in summer at the time of the monsoon, and is attributed to direct entrainment of stratospheric air during monsoonal cumulus cloud activity.

Mean <sup>36</sup>Cl/Cl ratios generally increase with distance from the coast. These are governed by stable Cl concentrations which decrease with distance from the coast with a dependence which is well-described by two exponentials. Comparisons with previous stable chloride measurements from Australia, however, demonstrate this relationship to have a significant geographic variability.

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**ION SOURCE DESIGN CRITERIA FOR AMS***L. R. KILIUS,<sup>1</sup> A. E. LITHERLAND, X.-L. ZHAO, R. P. BEUKENS and W. E. KIESER*

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A variety of ion sources is being designed for AMS and these include both negative and positive ion sources. The emphasis is usually on high ion currents because of the high isotope ratios, stable and reproducible operation for accurate measurements, rapid sample changing to maximize throughput, and the minimization of memory effects. These requirements are difficult to fulfil for moderate accuracy (<1%) measurements and very difficult for high accuracy measurements (<0.1%). Some recent contributions from the IsoTrace Laboratory towards these goals will be discussed.

<sup>1</sup>Deceased January 1996**MOLECULAR FRAGMENT PROBLEMS IN HEAVY ELEMENT AMS***L. R. KILIUS,<sup>1</sup> X.-L. ZHAO, A. E. LITHERLAND and K. H. PURSER*

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Heavy element AMS is made more difficult by the presence of the negative ion fragments of mass and charge (m,q) which come from molecules with the same mass as the heavy element M. If the heavy element after atomic or molecular dissociation has charge Q then a useful quantity is the integer n given by  $n = |Mq - mQ|$ . When  $n = 0$ , a rare heavy element is accompanied by strong beams of lighter molecular fragments (pilot beams) which usually prohibit the measurement of the rare species. The same problem occurs for  $n = 1$  because molecular beams undergo Coulomb explosion even in a gaseous target and the energy spread results in the increase of the resolution requirements of the high energy electric analyzers. The resolution needed for both velocity and electric analyzers, assuming that a higher resolution magnetic analyzer is also used, is given by  $mQ/n$ . In the case of  $^{129}\text{I}^{+4}$  analysis,  $^{97}\text{Mo}^{+3}$  interference and the add up from the other fragment can overwhelm the rare ions, requiring charge +5 to be used. Fortunately  $^{103}\text{Rh}^{+4}$  is very rare. Strategies for solving this heavy element AMS problem and some recent results will be discussed.

<sup>1</sup>Deceased January 1996**DETERMINATION OF RADIONUCLIDE PROTON CROSS SECTIONS ON THIN FOIL TARGET MATERIAL – IRRADIATION DETAILS, CHEMICAL SEPARATION TECHNIQUES, AND AMS MEASUREMENTS***K. J. KIM,<sup>1</sup> A. BEVERDING,<sup>1</sup> P. A. J. ENGLERT,<sup>1</sup> J. M. SISTERSON,<sup>2</sup> M. W. CAFFEE,<sup>3</sup> C. CASTANEDA,<sup>4</sup> J. VINCENT<sup>5</sup> and R. C. REEDY<sup>6</sup>*

Protons are the major constituent of galactic and solar cosmic rays (GCR and SCR, respectively). These protons have produced radionuclides in terrestrial and extraterrestrial materials since the formation of our solar system. Although both the GCRs and SCRs are dominated by protons, their energies are considerably distinct: SCR protons typically possess <200-MeV energy while typical GCR proton energies are 0.1–10 GeV. The long-lived radionuclides  $^{10}\text{Be}$  and  $^{26}\text{Al}$  are especially important because their half-lives span geologically and cosmologically interesting and significant time periods. Studies of cosmogenic nuclides such as  $^{10}\text{Be}$  and  $^{26}\text{Al}$  and their applications in studies of extraterrestrial and terrestrial materials have been hampered by the lack of reliable cross-section

measurements. Proton irradiation, chemical separation, and accelerator mass spectrometry (AMS) are the three major experimental components in the determination of cross sections.

To obtain optimum precision in the cross-section value for each element, the proton flux in each irradiation procedure should be properly monitored and isobaric interferences in chemical separation procedure should be minimized. The target materials Al, C, Mg, Si, and SiO<sub>2</sub> were irradiated with protons whose energies ranged from the threshold for the reaction of interest to 500 MeV. Target and proton-energy selections were based on the scarcity of proton cross-section data. Proton irradiations were performed at the University of California, Davis, Crocker Nuclear Laboratory cyclotron ( $E_p < 67.5$  MeV), Harvard Cyclotron Laboratory ( $60 < E_p < 160$  MeV), and TRIUMF ( $250 < E_p < 500$  MeV). The proton energy loss in each target was calculated using Monte Carlo techniques. The beam current was measured with a Faraday cup and compared to <sup>22</sup>Na production in aluminum monitor foils. At lower energies, below reaction thresholds, a secondary electron emission monitor was utilized.

Carriers of Al and Be were added at the beginning of the chemistry procedure for each of the targets. After dissolution of the target material, the Al and Be from each target were then chemically separated by ion chromatographic techniques. For the AMS measurement of <sup>10</sup>Be, it is critical that interferences from <sup>10</sup>B be minimized. Repeated purification before and after cation exchange procedures provided significant reduction of boron contamination in these samples. Approximately 70 targets were irradiated for <sup>10</sup>Be and <sup>26</sup>Al cross-section measurements. Because of radioactivity from short-lived radionuclides, it is necessary to wait 3 yr before sample preparation can begin. At the present time over half of the targets prepared for <sup>10</sup>Be and <sup>26</sup>Al measurements have been measured by AMS. The <sup>26</sup>Al/<sup>27</sup>Al ratios vary from  $1 \times 10^{-9}$  to  $1 \times 10^{-14}$  and the <sup>10</sup>Be/<sup>9</sup>Be ratios range from  $1 \times 10^{-12}$  to  $1 \times 10^{-14}$ . The errors in the AMS measurements are relatively small, so reasonably accurate excitation functions for each element can be obtained. It was observed that using accelerator mass spectrometry, the detection limit can be reduced down to  $10^5$  of <sup>10</sup>Be atoms in a sample.

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### GASTROINTESTINAL ABSORPTION OF ALUMINIUM IN PATIENTS WITH ALZHEIMER'S DISEASE MEASURED BY <sup>26</sup>Al ACCELERATOR MASS SPECTROMETRY

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The possible implication of aluminium in the aetiology of the senile dementia known as Alzheimer's Disease (AD) is controversial. Whilst it appears to be firmly established that aluminium concentrations in Alzheimer brain tissue are elevated relative to healthy controls, and that at the microscopic level aluminium is located particularly in senile plaques and neurofibrillary tangles (characteristic lesions of the disease), it is not established whether the aluminium is a cause or a result of the condition, or even merely a spectator. Previously, we have shown that gastrointestinal absorption of aluminium is enhanced in Down's Syndrome, a condition in which some of the neuropathological features (especially plaques and tangles) of AD appear by early middle age, and which (in common

with AD) may reflect an expression of the APP (amyloid precursor protein) gene located on chromosome 21. Noting the possible connections, we have now investigated aluminium uptake in Alzheimer patients.

Following ethical approval, a total of 13 patients and 13 control subjects were investigated. Subjects were given a citrate-containing drink labeled with  $^{26}\text{Al}$  (20ng, with  $20\mu\text{g}$   $^{27}\text{Al}$  carrier), and blood taken at 1 hr. Plasma was spiked with  $^{27}\text{Al}$ , converted to  $\text{Al}_2\text{O}_3$ , and the  $^{26}\text{Al}:^{27}\text{Al}$  ratio determined by AMS using the 14UD accelerator at the Australian National University. Aluminium uptake was estimated by assuming a patient plasma volume of 3 l. Whilst we have shown previously that the plasma-Al concentration under these conditions is determined by a number of kinetically controlled mechanisms, and are therefore not able to give an absolute measure of uptake, we argue that inter-comparison of the values obtained in the manner described for the members of the investigated groups should allow reliable values for relative uptakes to be derived. On this basis, we find that, compared to the control group (who show little within-group variation), the AD group show a wide variation in uptake, ranging upwards from the average value shown by the controls to *ca.* 5 times this value. Although means of the two groups are not significantly different at the 5% probability level (although they are distinguished at the 10% level), it seems possible that the AD group comprises two sub-groups, one of which shows markedly different behavior to the controls (to test this hypothesis will require a study on *ca.* 10 times the present scale).

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### THE INFLUENCE OF DISSOLVED SILICATE ON THE PHYSIOLOGICAL CHEMISTRY OF ALUMINIUM, STUDIED IN HUMANS USING TRACER $^{26}\text{Al}$ AND ACCELERATOR MASS SPECTROMETRY

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Aluminium is now recognized as a toxic element and control of human exposure is seen as a desirable public health objective. Uptake of ingested aluminium is known to be affected by chemical speciation, and it has been postulated that interaction with silicate could be a major factor affecting gastrointestinal availability. Additionally, silicate could affect aluminium physiology, for example cellular uptake, tissue storage and renal handling.

These hypotheses have now been tested in two human subjects. Blood aluminium concentrations and urinary excretion have been measured at frequent intervals for 7 days following ingestion of an  $^{26}\text{Al}$ -labeled Al dosing solution (20 ng in  $20\mu\text{g}$ , respectively), both with and without added silicate (2 mg, also in solution). Blood plasma was separated into low and high molecular size fractions by membrane ultrafiltration at 5 kD cut-off. Biological samples were spiked with  $^{27}\text{Al}$ , converted to  $\text{Al}_2\text{O}_3$ , and the  $^{26}\text{Al}:^{27}\text{Al}$  ratio determined by AMS using the 14UD accelerator at the Australian National University. The measured isotope ratio ranged from 10–9 to 10–13, the effective baseline in these experiments being ~10–14. The tracer concentration was used to derive the total Al intake, uptake, retention and excretion as a function of time, using a simple 3-compartment kinetic model. Major differences were observed in Al behavior in the absence and presence of silicate. In the former case, plasma-Al concentrations passed through a single maximum *ca.* 1 hr after dosing, *ca.* 5% of the plasma-Al was ultrafilterable, and renal excretion could be quantified by one clearance parameter (~5% nominal maximum clearance, perhaps corresponding to the ultrafilterable fraction).

In the latter case, plasma-Al concentrations pass through two distinct maxima, the later one apparently corresponding to that observed without silicate, and renal excretion was initially higher than that observed previously. We suggest that silicate interacts chemically with Al in solution, and that talumino-silicate complex so formed is both absorbed and excreted more rapidly than non-(Si)-complexed Al.

These observations could be relevant to the interpretation of epidemiological studies of the incidence of Alzheimer's disease, which has been correlated with Al concentrations in drinking water, as dissolved silicate could act as a confounding factor in those studies. There are also public health implications if aluminium uptake and retention is affected by silicate concentrations in drinking water supplies.

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## RADIOCARBON DATING ORGANIC RESIDUES AT THE MICROGRAM LEVEL

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Previous studies have determined that the total system <sup>14</sup>C background in AMS systems for catalytically-reduced graphitic carbon samples below *ca.* 500 μg are inversely proportional to their weights (Vogel, Nelson and Southon 1987). We have examined this relationship down to 10 μg using assumed <sup>14</sup>C-“dead” blanks (purported Pliocene wood) and to 40 μg using contemporary standards (ANU sucrose) (Kirner, Taylor and Southon 1995). Our observations are consistent with Vogel, Nelson and Southon (1987) with respect to the inverse relationship between sample weight and <sup>14</sup>C activity. The data indicate a constant addition of modern contamination during the graphitization process resulting in the observed background <sup>14</sup>C activity in graphitic carbon samples. This report will focus on our current studies, which include the differential effects on background and contemporary standards using cobalt and iron as catalysts and the results of the AMS <sup>14</sup>C dating of microgram amounts of amino acids extracted from fossil bone.

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## INVESTIGATIONS OF THE HUMAN ALUMINUM BIOKINETICS WITH $^{26}\text{Al}$ AND AMS

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Aluminum biokinetics in humans was studied by administering oral and intravenous doses of  $^{26}\text{Al}$  to four healthy volunteers of ages between 21 and 54 yr and two patients with renal failure of ages 54 and 57 yr. The oral dose was 100 ng  $^{26}\text{Al}$  together with 100 5g Al as  $\text{AlCl}_3$ . The IV dose was 1 ng  $^{26}\text{Al}$  together with 20 5g Al, also as  $\text{AlCl}_3$ . The administered doses were far from saturation values. The radiation energy doses calculated for the whole body with the compartment model to be described were 0.1 and 0.7 5Sv, respectively. Blood samples were taken at times between 20 min and half a year after administering the doses. The daily urine was collected in the first nine days, spontaneous urine samples were taken at later times when the blood samples were obtained. Creatinin renal clearances and haematocrite values were measured in the time period of the investigation as well. After addition of Al carrier and physical and chemical treatment of the samples was taken,  $^{26}\text{Al}$  was measured with the München Tandem accelerator using the Wein velocity filter and a Bragg ionization chamber. The detection limit for  $^{26}\text{Al}/\text{Al}$  was  $<10^{-15}$ .

For the IV-administered doses, and after one day for the doses administered orally to the healthy volunteers, a roughly linear dependence is observed between the logarithm of  $^{26}\text{Al}$  concentrations in serum and logarithm of time. This dependence is also observed in a human IV study by Day *et al.* (1994) over a period of three years. Both patients show time curves that are different than those of the healthy volunteers.

An open-compartment model was developed and used in order to describe the time dependences of the measured  $^{26}\text{Al}$  concentrations in blood and urine and also to establish the human Al biokinetics. The model comprises stomach and duodenum for oral administering, three central compartments for blood and interstitium (Al in interstitium) and three peripheral compartments that are needed to describe the time dependence for the long observation period of up to three years. Two peripheral compartments are connected with the protein-bound central Al compartment. The third peripheral compartment (interpreted as bones) is connected with the interstitium. The excretion occurs from the central compartment comprising the Al-soluble in serum into the urine.

From the collected daily urine samples, the Al resorption factors were determined to be in the range of 0.1% for all doses administered orally as  $\text{AlCl}_3$ . No differences are seen in the resorption factor between healthy volunteers and patients. By comparing creatinin and  $^{26}\text{Al}$  renal clearances, the amount of Al soluble in serum was obtained. Time constants (*i.e.*, inverse-rate constants) between the compartments were derived. These time constants are in the range of 10 min to 1 yr. The model is able to reproduce the measured time dependences in blood and urine and also the amount of 4% of  $^{26}\text{Al}$  still observed in the body after three years of IV-administered  $^{26}\text{Al}$  (Day *et al.* 1994). In the model, this 4% is found to be bound to the bones. For the patients, the sizes of the two peripheral compartments connected to the central compartment of the protein-bound Al are obtained to be different as compared to the healthy volunteers. Al compartment sizes are the maximum number of Al places available and to be occupied in a compartment. These sizes are given by the ratios of the time constants.

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## ENRICHMENT OF SUB-MILLIGRAM SIZE CARBON SAMPLES

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Enrichment of carbon isotopes has been developed in the past for conventional dating to extend the radiocarbon time scale beyond 50 ka. For AMS, the necessary system can be scaled down considerably. We have developed an enrichment system for use in conjunction with the Groningen AMS. Our goal is to increase the <sup>14</sup>C count rate for very small (even for AMS) samples.

We enriched samples in the form of CO by thermodiffusion. We built a column with a height of 3 m, with a central hot wire with a temperature of ca. 900 K. The enriched CO is collected in a reservoir connected to a graphitization cell. For <sup>13</sup>C we obtained an enrichment of about a factor of 3, depending on various parameters. Consequently, we expect for <sup>14</sup>C an enrichment of about a factor of 6. Preliminary AMS measurements will be presented at the conference.

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## AMS AT THE MUNICH GAS-FILLED ANALYZING MAGNET SYSTEM GAMS

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For AMS measurements with heavy isotopes (e.g., <sup>59</sup>Ni), the deflection of the ions by a gas-filled magnet is a very effective and efficient method to suppress isobaric background. Interacting with the gas atoms, the ions get an average magnetic rigidity depending on their nuclear charge. Measurements were done at Argonne National Laboratory (Paul *et al.* 1989) with a Split Pole and in Munich (Korschinek *et al.* 1994) with a Q3D-spectrograph. The weak points of these systems are field arrangements, where only 40–50% of the ion paths in the gas-filled regions are covered by magnetic field. Therefore, a simple bending magnet should reveal optimum conditions. Here nearly the total length of the trajectories in the gas is covered by magnetic field, so the statistics of the charge state fluctuations improve, while the small-angle scattering is unchanged. For these reasons we have set up a dedicated gas-filled magnet system GAMS ( $\varphi = 135^\circ$ ,  $R = 90$  cm). The first and the third of the detector's four energy loss sections are split to obtain the position and angle of the incoming ions. For additional suppression of isotopic background a 3.5 m time-of-flight path has been installed. First measurements of the nuclides <sup>36</sup>Cl, <sup>53</sup>Mn, <sup>59</sup>Ni and <sup>60</sup>Fe have been performed. Background limits achieved here were  $^{59}\text{Ni}/\text{Ni} < 1 \times 10^{-13}$ ,  $^{60}\text{Fe}/\text{Fe} < 1 \times 10^{-14}$  and  $^{36}\text{Cl}/\text{Cl} < 3 \times 10^{-15}$ .

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## ONE TO 200 AMU TUNABLE PRETZEL MAGNET NOTCH-MASS-FILTER AND INJECTOR FOR TRACE ELEMENT ACCELERATOR MASS SPECTROMETRY

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A tunable Pretzel magnet notch-mass-filter and injector was developed to facilitate the study of trace element distributions in electronic and geologic materials using the Naval Research Laboratory (NRL) trace element accelerator mass spectrometer (TEAMS) (Grabowski *et al.* 1996). The high energy beam transport system is capable of simultaneously analyzing a broad momentum range of  $p_{\max}/p_{\min} \approx 2.8$  ( $M_{\max}/M_{\min} \approx 8$ ). Therefore, a wide-mass-band injection system had to be developed to fully utilize the high energy capabilities.

A Pretzel magnet (Enge 1963) (magnetic mirror) with a field index  $n$  of 0.925 and a loop of  $264.6^\circ$  provides beam optical properties that are ideal to use as a mass-filter for the injection system. Its optical properties are basically parallel to point to parallel in both planes. All trajectories have the same shape, but loop size is dependent on momentum. The penetration depth along the mirror plane scales with particle momentum as  $\sim(\text{momentum})^{1/(n+1)}$ . The NRL design has a 70-cm-long mirror plane with a flag system built into slots in both of the poles for positioning up to 8 beam masks with  $\approx 1$  mm resolution. The masks will be used to block intense matrix-related beams (*e.g.*, Si, SiO, Si<sub>3</sub>, *etc.*) while passing other less intense beams of more interest into the accelerator. Otherwise, charge exchange and multiple scattering events from the intense beams would introduce background and mask interesting parts of the mass spectra. The mass filter will have near-amu mass resolution over the range of 1 to 200 amu at 40 keV.

The primary and secondary columns of a Physical Electronics model 6300 quadrupole secondary ion mass spectrometer (SIMS) form the key components of the negative ion source for the injection system. These commercially proven components will facilitate depth profiling, imaging, and the analysis of both conducting and insulating substrates at pressures as low as  $10^{-10}$  torr. The secondary column is equipped with a  $90^\circ$  spherical electrostatic analyzer (ESA) with an energy resolution of  $E/\Delta E$  of 25. Because the maximum beam energy of the secondary column is only 350 eV, the beam will be accelerated to 6 keV once it leaves the column by a low-aberration double gap lens. This lens will focus the beam to the focal position of a 40-kV accelerator tube, thus providing the required parallel beam to the subsequent Pretzel magnet notch-mass-filter. After leaving the Pretzel, the parallel beam will be focused to the object position of the accelerator using a high conductance Einzel lens. This will keep the base pressure in the first accelerator tube in the  $10^{-8}$  torr range. Further detail of the injector will be presented.

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## CURRENT STATUS OF THE AMS SYSTEM AT THE UNIVERSITY OF TOKYO

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The tandem accelerator facility constructed between 1991 and 1994 at the University of Tokyo was designed with special emphasis on micro-analytical studies using techniques like AMS, PIXE, RBS, NRA and nuclear microprobe analysis, and on researches in atomic and nuclear physics. The new facility is equipped with a 5 MV tandem accelerator, two ion sources for gas and solid materials (the latter one is a multi-cathode Cs sputter type used mainly for AMS), nine beam lines dedicated to micro-analytical researches and many special items designed for AMS experiments. The facility is called MALT, which stands for "Micro Analysis Laboratory, Tandem accelerator". During the last year we struggled against many initial failures occurring both in hard- and software and *ca.* 40% of the machine time has been used for AMS programs including research and development.

The largest problem still remained unsolved is the low beam transmission through the accelerator that might be caused by wrong alignment or by squeezed setting of the optical elements. For the case of <sup>14</sup>C measurements with 5 MV terminal voltage, the best particle transmission we have obtained so far was *ca.* 50%. Similar values were also measured for <sup>10</sup>Be and <sup>26</sup>Al, which affects the reproducibility, sensitivity and accuracy of all the measurements performed with the sequential injection method (jumping method). This is not the case for the measurements carried out using the internal beam monitor method that is not influenced by the wrong beam transmission. In the table is shown the performance of the MALT-AMS in the initial developing stage.

For the near future we plan to carefully investigate the origin of the background and to extend the AMS measurements to other radioisotopes. For the <sup>36</sup>Cl measurement a gas-filled magnet is presently under development. In addition, a TOF system, which will be used for <sup>129</sup>I-AMS, has been already installed and will be tested soon.

Nuclide	AMS method	Typical ion current from ion source; mA	Accuracy %	Background X*/X =
<sup>10</sup> Be	Beam monitor	BeO-; 1	<2	< 5 × 10 <sup>-14</sup>
<sup>14</sup> C	Beam monitor	C-; 30 (graphite)	<2	< 1 × 10 <sup>-15</sup>
	Sequential injection	C-; 5 (amorphous)	3-4	< 3 × 10 <sup>-15</sup>
<sup>26</sup> Al	Sequential injection	Al-; 1	3-4	< 1 × 10 <sup>-13</sup>

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**LONG-TERM VARIATIONS OF SOLAR INTERNAL AND EXTERNAL ACTIVITY***G. E. KOCHAROV and O. G. GLADYSHEVA*

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We discuss the status and potential of studying solar flare and modulation activity, solar neutrino flux over long-time scales by high-precision and sensitivity measurements of cosmogenic isotope content in natural archives of cosmic rays and long-lived nuclides in Earth's crust that were produced by solar neutrinos.

*1. What is the nature of the global deficit in solar neutrinos?*

In all four series of experimental data there is a deficit in solar neutrino fluxes in comparison with the prediction of standard solar models (SSM). Such a general deficit was predicted by Kocharov and Starbunov in 1969 based on the hypothesis of increased abundance of  $^3\text{He}$  in the deep solar interior. To explain the data obtained, the  $^3\text{He}$  abundance has to be  $3 \times 10^{-5}$ , which is several times more than SSM prediction. Such an increased abundance is possible: if the primordial Sun already had helium-3 or sudden mixing may have occurred at some time in the solar interior (the idea of W. Fowler, 1972). In the last case, the Sun now should be in a transition region. The forthcoming experiments of Superkamiokande and Borexino will answer the questions formulated above. In the case of a positive answer, geochemical experiments based on the reactions  $^{98}\text{Mo}(\nu_e, e^-)^{98}\text{Tc}$ ,  $^{97}\text{Mo}(\nu_e, e^-)^{97}\text{Tc}$  become the first priority.

*2. What is the highest possible total energy of solar flare accelerated particles?*

Available experimental data show that the higher the total energy of solar cosmic rays, the lower the observation probability. Therefore, to get maximum value we need measurements on a long-time scale. At the moment, there is only one possibility, connected with high-precision measurements of nitrate abundance in polar ice and  $^{14}\text{C}$  abundance in the natural archives of cosmic rays: tree rings, ice cores, corals, etc.

*3. What is the nature of deep solar minima?*

High-precision year-by-year measurements of  $^{14}\text{C}$  abundance in tree rings first allowed the reconstruction of GCR intensity before, during, and after deep solar minimum (Maunder). It is shown that even during deep minimum GCR modulated by the Sun. Extremely important are year-by-year measurements of  $^{14}\text{C}$  abundance in the tree rings for the Sporer minimum (AD 1416–1534) of solar activity. Based on the data for two minima with high probability, the problem of solar modulation of GCR during deep minima has to be solved.

 **$^{60}\text{Fe}$ , A PROMISING AMS ISOTOPE FOR MANY APPLICATIONS***G. KORSCHINEK, T. FAESTERMANN, K. KNIE and C. SCHMIDT*

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$^{60}\text{Fe}$  ( $T_{1/2}=1.5$  My) (Kutschera *et al.* 1984) has been considered as an AMS isotope in only a few cases: once for measurements in meteoritic studies, due to its production in spallation reactions in iron meteorites by galactic cosmic rays (Paul *et al.* 1989); it has also been recognized applications in medicine (Paul and Korschinek 1989).

Using our dedicated gas-filled GAMS (Knie *et al.* this conference), we have studied some applications of  $^{60}\text{Fe}$ . Three major fields have been identified:

1. *Medicine*—Studies of the iron metabolism in humans at extremely low specific radioactivity. Additional use of another long-lived radioisotope,  $^{55}\text{Fe}$  ( $T_{1/2}=2.7$  yr), also detectable by AMS (Müller *et al.* 1990), allows even double tracer studies with low specific radioactivity.
2. *Nuclear Physics*—The mass distributions of fission products from neutron-induced fission have been measured already down to about the mass 68. In order to look for superasymmetric fission, the extremely sensitive detection of  $^{60}\text{Fe}$  in fission products opens the possibility of testing present fission theories.
3. *Astrophysics*—Type II supernovae are expected to be the main sources for  $^{60}\text{Fe}$  distributed in the interstellar medium. By detecting the  $^{60}\text{Fe}$  concentration in sediments on Earth, one could get the first positive measurement of  $^{60}\text{Fe}$  abundance in the interstellar medium and thus information on the nucleosynthesis in supernovae and the galactic transport of matter released by supernovae.

Type II supernovae are expected to be the main sources for  $^{60}\text{Fe}$  distributed in the interstellar medium. By detecting the  $^{60}\text{Fe}$  concentration in sediments on earth, one could get the first positive measurement of the  $^{60}\text{Fe}$  abundance in the interstellar medium and thus information on nucleosynthesis in supernovae and the galactic transport of matter released by supernovae. For all these applications, sensitivities down to a level of  $10^{-14}$  are needed. We have studied the GAMS for measurements of  $^{60}\text{Fe}$ . For this purpose, a calibration sample with a concentration of  $9 \times 10^{-12}$  has been produced. First measurements and the achieved sensitivities will be presented.

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#### THE REDUCTION OF SAMPLE MEMORY EFFECTS IN THE CHALK RIVER AMS ION SOURCE

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At Chalk River  $^{36}\text{Cl}/\text{Cl}$  or  $^{129}\text{I}/\text{I}$  ratio determinations of samples collected from nuclear facility environments have, on occasion, been compromised by sample-to-sample interference from residue within the ion source. This limits the dynamic range of ratio measurements to a factor of 500 and often leads to excessive downtime for ion-source refurbishing. In order to understand and reduce ion source memory effects, the distribution of sputtered material has been measured by radioactive tracer techniques. In addition, *in-situ* measurements by elastic recoil detection on an operating ion source have identified the levels of surface contaminants as a function of surface temperature. These measurements indicate that the remembered ions originate from a region immediately surrounding the sample. Memory effects can be mitigated by operating this portion of the ion source above  $350^\circ\text{C}$  and by screening this region through careful sample-holder design. The presentation will describe these measurements and our findings.

**THE CHALK RIVER AMS TIME-OF-FLIGHT DETECTION SYSTEM**

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At Chalk River the final detection and identification of  $^{129}\text{I}$  is performed through time-of-flight (TOF) measurements over a 2.5-m path following rigidity and velocity analyses. The start/stop signals are derived from microchannel plates that detect accelerated secondary electrons emitted from thin carbon films. Because velocity straggling affects the time resolution and hence the ability of a TOF system to discriminate against neighboring mass contaminants, the Chalk River start detector has been constructed so that the particle beam is unobstructed by grids. It provides a resolution of 400 ps (FWHM) for the 50 MeV  $5^+$  iodine ions. The TOF peaks are gaussian to the 0.0002 level and masses 128 and 130 are separated from  $^{129}\text{I}$  by *ca.* 2 ns.

In addition to TOF, the total energy (E) of the ions is measured with a Si surface barrier detector. Although  $^{129}\text{I}$  standards and NaI blanks require only TOF for unambiguous identification of  $^{129}\text{I}$ , many environmental samples are characterized by a plethora of peaks where both TOF and E are essential for unambiguous identification of  $^{129}\text{I}$ . This presentation will provide details of the apparatus and our operating experience.

[THE CHALK RIVER AMS SAMPLE CHANGER AND ION SOURCE, Koslowsky *et al.*: see p. 134]

**THE ERLANGEN AMS FACILITY: STATUS REPORT AND RESEARCH PROGRAM**

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The Erlangen AMS facility (Arslan *et al.* 1994) consists of a high current sputter ion source, a  $90^\circ$  injection magnet with fast isotope switching, an EN tandem accelerator, a  $15^\circ$  electrostatic deflector, a  $55^\circ$  analyzing magnet (for  $^{14}\text{C}$ ), a  $120^\circ$  magnetic split pole spectrometer (for the heavier ions), and a  $\Delta E$ -E gas detector. The sputter ion source (Höpfl *et al.* 1992) equipped with a spherical ionizer and a cassette for 50 samples is fully computer controlled and delivers  $^{12}\text{C}^-$  currents of up to  $150\mu\text{A}$  at an energy of 55keV. Improvements of the vacuum situation within the ion source ( $10^{-6}$  mbar), the low and high energy beam line ( $5 \times 10^{-7}$  mbar) led to a strongly improved particle transmission through the facility of up to 80% taking into account the charge distribution in the high voltage terminal. Thus even with conservative ion source conditions with  $25\mu\text{A}$   $^{12}\text{C}^-$  current a  $^{14}\text{C}$  count rate of 100Hz can be obtained for an ANU calibration sample. Another consequence of this improved vacuum was a drastic reduction of the machine background determined with graphite samples which are measured with 0.07 PMC  $^{14}\text{C}$ -content corresponding to an apparent age of 58,000 yr.

In the last year, efforts have been made to optimize the sample preparation for the sputter targets producing high  $^{12}\text{C}^-$  currents independent of the original sample material. For sediment- and tree-samples the usual acid-alkali-acid pretreatment is performed. The remaining material is oxidized by heating it to  $900^\circ\text{C}$  under vacuum in a closed quartz glass tube together with copper oxide and silver wool. Then graphitization takes place by heating a mixture of  $\text{CO}_2$  and  $\text{H}_2$  at  $625^\circ\text{C}$  with iron powder as a catalyst. The  $^{14}\text{C}$  background induced by this chemical processing has been measured to be  $<0.7$  PMC from graphite samples which were treated in the same way. For very small samples containing less than *ca.*  $100\mu\text{g}$  carbon a new reduction facility with a total volume of only  $3.8\text{ cm}^3$  has been built recently. This will enable AMS measurements of pollen and environmental air samples.

In the routine sequence of AMS measurements the quality of the whole facility is first checked by calibrated samples (ANU, wood from 1860 AD, 23 PMC IAEA, graphite) which have been chemically treated in the same manner as the unknown samples. Then 2–3 unknown samples are measured before another calibrated sample is used. In this way accuracies of better than 1% have been established.

At the moment, the research program involves only  $^{14}\text{C}$  measurements, but after the installation of a new injection system consisting of a  $90^\circ$  electrostatic deflector followed by a new stronger  $90^\circ$  magnet applications with heavier elements will be pursued. Now the interest is mainly focused on the dating of sediment profiles from Bavaria. In addition, several archaeological samples and volcanic samples have been dated. A new project starting this year involves  $^{14}\text{C}$  measurements on chlorine hydrocarbons. The aim of this project is to deduce the origin of these compounds, *e.g.*, natural or anthropogenic, from the measured  $^{14}\text{C}$  content.

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#### $^{14}\text{C}$ DATING OF SEDIMENT SAMPLES

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The research program at the Erlangen AMS facility (Kretschmer 1996) is mainly concentrated to the dating of sediment samples. Recently we investigated some peat profiles from South Bavaria as part of the program "Change of the Geo- and Biosphere During the Last 15,000 Years, Continental Sediments as Expression of Changing Environment." The dating of several continuous peat profiles at different locations together with corresponding pollen analyses should help to establish a better chronology of climate and vegetation of Holocene in Bavaria.

First, bulk sediment samples were obtained from the core by AAA treatment, conversion to  $\text{CO}_2$  under vacuum and finally  $\text{H}_2$  induced graphitization with the use of iron as a catalyst. One problem with the radiocarbon dating of peat samples is the "hard water effect" since the plants on which the peat is based can assimilate  $\text{CO}_2$  both from the air and from the water. If old carbonates are dissolved in the water the dating of the bulk sediments may be obscured by this effect and the resulting ages become older. This problem can be solved if the dating is performed on the pollen, which are blown in from outside. Therefore we plan pollen dating to obtain more reliable results. For the extraction of the pollen we use a filter with a  $100\ \mu\text{m}$  nylon mesh; besides the pollen the filtrate contains also silicates, amorphous organic material and cellulose, which are removed by chemical treatment described in Faegri and Iversen (1989), and sieving through a  $20\text{-}\mu\text{m}$  nylon mesh. The result of this procedure is checked under microscope and converted first into  $\text{CO}_2$  and then via graphitization into sputter targets. First pollen dating has been performed and the results are encouraging.

Another point of interest was the eruptive chronology of the Sakurajima volcano in southern Kyushu, which is one of the most active volcanoes in Japan. Tephra layers of large-scale eruptions of this volcano may serve as time markers in this region and have been investigated extensively (Machida and Arai 1996). Since the oldest tephra layer is subject to a controversy concerning its

age, several samples of this layer have been dated with our AMS facility. Preliminary results of 25,000–26,000 yr BP confirm the results of Machida and Arai (1996) and imply that eruptive history of the Sakurajima volcano started at this time.

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## PRODUCTION RATES FOR EXPOSURE DATING: TO BE WITHIN THE RADIOCARBON CALIBRATION CURVE

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Sample preparation and the measurement of the cosmogenic nuclide concentration using ultrasensitive mass spectrometric methods such as accelerator mass spectrometry (AMS) are, under normal circumstances, not the limiting factors anymore for the accuracy of an exposure age or an erosion rate. The main contribution comes from the limited knowledge of the production rates of cosmogenic nuclides, both as a function of location and time. The uncertainty is estimated to be of the order of 10% for ages in the 10 kyr range (Nishiizumi *et al.* 1989) and quite unknown for time scales of the order of millions of years.

The production rates used today are generally based on radiocarbon ages of material found either on the rock surfaces (rock varnish) or somewhere nearby, and are associated with glacial events towards the end of the last glacial cycle (Nishiizumi *et al.* 1989), which are of course not dated in an absolute fashion. One can also expect an uncertain delay for the rock varnish to build on a fresh surface. In any case however, in principle <sup>14</sup>C ages can be converted to calendar ages using the <sup>14</sup>C calibration curve. This curve, however, extends undisputed only to *ca.* 10,000 BP. The difference between <sup>14</sup>C ages and calendar ages at *ca.* 10,000 BP is already *ca.* 1000 yr or *ca.* 10%. The samples used so far to determine production rates fall outside this range increasing the uncertainty. It would be very desirable to, as a first step, obtain production rates for events within the <sup>14</sup>C calibration curve.

We have identified such an event with known calendar age from calibrated <sup>14</sup>C dates, sampled rock surfaces and measured (so far only) <sup>10</sup>Be concentrations in them. This event was an enormous rock slide near Köfels (Austria) (Erismann *et al.* 1977), where *ca.* 2–3 km<sup>3</sup> of rock slid off a mountain and deposited material on the other side of the valley as high as 500m above the valley floor. The event has been dated to 8710 <sup>14</sup>C yr (Heuberger 1966) with tree material found in an exploratory mine shaft driven into the deposited material *ca.* 300 m above the valley floor. Additional tree material will be dated at the Zurich AMS facility to confirm the age. We have taken samples from the sliding surface (polished quartz planes) and samples from the crest of the toe of the landslide on the other side of the valley. Preliminary results are in the expected range. Shielding corrections for the surrounding mountains, estimates for shielding by forest vegetation and snow cover have still to be looked into

more accurately. We expect to have a series of rocks measured, and we plan to present the data in a form that allows discussion the various corrections made to the final result for a time integrated production rate for the last 10,000 yr.

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## AMS FACILITY AT NATIONAL INSTITUTE FOR ENVIRONMENTAL STUDIES

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A new AMS system based on a 5 MV tandem accelerator has been installed at National Institute for Environmental Studies (NIES), Tsukuba. This system, NIES-TERRA (Tandem accelerator for Environmental Research and Radiocarbon Analysis), is used primarily for precise measurement of <sup>14</sup>C/<sup>12</sup>C and <sup>13</sup>C/<sup>12</sup>C isotope ratios to investigate the origin and global circulation of greenhouse gases such as CO<sub>2</sub> and CH<sub>4</sub>. Our research program is also directed towards using <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, <sup>41</sup>Ca, and <sup>129</sup>I as both tracers and chronometers in environmental studies.

The construction of the total system was contracted with NEC. The accelerator is a horizontal tandem Pelletron (15SDH-2) whose charging system can deliver *ca.* 250  $\mu$ A to the high-voltage terminal. Negative ion injection is performed by two combinations of ion source and injector. One of the ion sources is a multiple cathode cesium sputter type for solid target (MC-SNICS). Up to 40 cathodes at a time can be held on the fluid cooled cathode ring. Negative ions produced in the source are accelerated to a maximum of 80 keV and fed into a fast bouncing sequential injector through a 45° spherical electrostatic analyzer (305-mm radius). This injector has a 90° double focusing magnet (ME/Z<sub>2</sub>=15, 457-mm radius) with a chamber insulated electrically from the ground to maintain the constant magnetic rigidity. Fast sequential injection of isotopes is performed by applying short high-voltage pulses to the insulated magnet chamber. The second ion source produces negative carbon ions by using cesium to sputter pure titanium surface onto which sample gas is continuously bled. Details of this multiple gas feed negative ion source (MGF-SNICS) are reported elsewhere in this conference. The MGF-SNICS is dedicated to accurate radiocarbon dating with a simultaneous isotope injector. In this injector, two magnets are used to separate and combine the negative ions of masses 12, 13, and 14 in the horizontal plane. Einzel lenses are provided to focus the ion beams into and out of the magnets. At the midpoint of the magnets, there is a slot lens for focusing three ion beams vertically. The rare radioisotopes are detected by a multiple electrode ion chamber. A time of flight setup with a path of 3 m can be used for a measurement of heavy isotopes.

Current status of our AMS facility will be reported.

**VERA: A NEW AMS FACILITY IN VIENNA**

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VERA is an acronym for Vienna Environmental Research Accelerator, a new center for accelerator mass spectrometry associated with the Institut für Radiumforschung und Kernphysik at the University of Vienna. The name of the facility indicates its primary mission, *i.e.*, the tracing of long-lived natural and artificial radionuclides in the five domains of our environment: atmosphere, biosphere, hydrosphere, cryosphere and lithosphere.

VERA is housed in a small two-story palace from the last century under landmark protection, whose interior was completely reconstructed to generate a modern laboratory for AMS. At the same time, efforts were made to preserve the original features of the building as much as possible. The total floor space available (~600 m<sup>2</sup>) was divided into four areas of roughly equal size: accelerator, mechanics and electronics shops, sample preparation, and offices including a seminar room. Particular emphasis was placed on providing ample space for sample preparation.

VERA is based on an AMS facility built by National Electrostatics Corporation in Wisconsin, USA. An important design feature is the capability of transporting ions up to the heaviest elements. The main components of VERA are a 40-sample Cs-beam sputter source, a 45-degree spherical electrostatic analyzer, a double-focusing 90-degree injection magnet with an insulated vacuum chamber for fast sequential isotope injection, a 3-MV pelletron tandem accelerator, a double focusing 90-degree analyzing magnet with a mass-energy product of 176 MeV amu, and a Wien Filter. Several different particle detector systems are under construction in Vienna. VERA is fully computer-controlled and integrated into the Internet, thus providing the possibility to interact with it from essentially any place on Earth.

VERA will initially be used to perform AMS measurements of <sup>10</sup>Be, <sup>14</sup>C, and <sup>26</sup>Al. In a second phase, suitable peripherals will be developed to utilize the full mass range of radionuclides. In this report, an overview of the facility with its current status and prospects for the future will be presented.

**RADIOCARBON AMS DATING OF THE SITES WITH EARLY POTTERY FROM THE RUSSIAN FAR EAST**

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We recently obtained radiocarbon AMS dates from two key sites in the Russian Far East, lower Amur River basin, Gasya and Khummi. They both contain the earliest remains of archaeological pottery, and belong to the Osipovka culture of Incipient Neolithic.

At the Gasya site (48°45'N, 135°40'E), charcoal collected from the hearth is associated with pottery (depth 2.1–2.4 m below surface) that was dated using conventional decay counting to 12,960 ± 120 BP, LE-1781. The other charcoal sample (depth 1.4–1.6 m) was AMS dated to 10,875 ± 90 BP, AA-13393.

At the Khummi site (50°34'N, 137°06'E), a conventional <sup>14</sup>C date associated with the upper level of the Osipovka stratum is 7,760 ± 120 BP, GIN-6945. Another charcoal sample from this level was AMS dated to 10,345 ± 110 BP, AA-13391. The second AMS date from the lower level of the Osi-



povka stratum is  $13,260 \pm 100$  BP, AA-13392. Charcoal collected without close association with artifacts and located 1 m away from any artifacts was AMS dated to  $42,800 \pm 1,900$  BP, AA-13394.

These new  $^{14}\text{C}$  AMS dates allow us to suggest that the appearance of pottery in the Incipient Neolithic of the lower Amur River basin could be as early as *ca.* 13,000–13,200 BP. Both the Gasya and Khummi sites have similar  $^{14}\text{C}$  dates to the Fukui Cave and Kamikuriowa site in southern Japan (12,200–12,700 BP). Thus, the pottery appeared in the Russian Far East at a similar time to that in the southern Japanese Islands.

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## RADIOCARBON IN MARINE DISSOLVED ORGANIC MATTER

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Dissolved organic carbon is the largest active reservoir of reduced carbon on Earth. It is defined as all organic material in water that passes a 0.2 micrometer filter. Part of the DOC is biologically active (young) and plays an important role in the ecology of surface waters as food source for bacteria (the microbial loop) and in the distribution of nutrients and carbon in the water column. Another part of DOC appears to be very refractive (old) and is present as a background for prolonged periods.

Analytically it is a very difficult property to measure because of its high chemical stability and its low concentration and consequent blank problems. We have developed a new method for oxidation of DOC and the recovery of the  $\text{CO}_2$  for isotopic measurement. We use the supercritical phase as oxidation medium to ensure a complete oxidation and prevent problems with salt precipitation. This method is used for isotopic measurement of the DOC ( $^{13}\text{C}$ ,  $^{14}\text{C}$ ). Knowledge of the  $^{14}\text{C}$  content gives insight into the relation between old and young DOC and the dynamics of the old pool. At the conference this method and its first results will be presented.

## FIELD VARIABILITY OF CARBON ISOTOPES IN SOIL ORGANIC CARBON

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The question of homogeneity of carbon isotopic composition of soil organic carbon becomes critical when trying to obtain some representative value for a site, and especially when attempting to quantify small changes in the isotopic composition in these carbon pools over time. Several years of free-air  $\text{CO}_2$  enrichment (FACE) experimentation have been done at the Maricopa Agricultural Center of The University of Arizona, *ca.* 50 km south of Phoenix. Among specific investigations, the petroleum-derived  $\text{CO}_2$  ( $^{14}\text{C}$ - and  $^{13}\text{C}$ -depleted) used to enrich the FACE plots in cotton and wheat experiments was used as an isotopic tracer to follow atmospheric carbon into the plants and then into the soils. The early work (Leavitt *et al.* 1994) with cotton indicated an isotopic shift in  $\delta^{13}\text{C}$  of the FACE

soils consistent with *ca.* 10% input of freshly derived carbon during the experiment, whereas TAMS-measured  $^{14}\text{C}$  activity produced some inconsistencies which prevented independent confirmation of the 10% input figure. The first few  $\delta^{13}\text{C}$  numbers also actually produced initial ambiguous results, but the weight of additional measurements produced a clearer picture. We hypothesized that some combination of lab precision and field variability may have contributed to uncertainty with few initial data points. In this study, soil was sampled from the location of eight new circular plots (23-m-diameter plots embedded in a *ca.* 9 ha farm field) just prior to the beginning of 1995–1996 FACE experiments with wheat. Each plot was sampled with at least four soil cores taken along equally spaced radii from the center and at depths of both 0–30 cm and 30–60 cm; in the case of two of the plots, 8 cores were taken. Soil samples were acidified with 1N HCl to remove soil carbonates, and plant fragments were removed by skimming from the liquid surface and by manual removal from dried, ground soil using forceps under 20 $\times$  magnification. Multiple combustion and analyses of the same soil sample indicate precision of *ca.* 0.3‰. The  $\delta^{13}\text{C}$  results on this total organic carbon fraction produced a range of average values among the plots of *ca.* 4‰ for the 0–30 cm depth and *ca.* 1‰ at the 30–60 cm depth. Within plots, the  $\delta^{13}\text{C}$  range was as much as 3‰ for both 0–30 cm and 30–60 cm soils, although the standard deviations for the deeper soils were in all cases smaller than for the surface soils. The results indicate substantial variability in the isotopic composition of the soil organic carbon, especially in the surface layer, in spite of the fact this layer has been heavily plowed over several decades of farming. These fields were leveled early in their history, which could account for some of the variability depending upon specific removal and additions of soils to different areas in the field. Our results demonstrate the need for sufficient subsampling to characterize the isotopic composition of this organic carbon pool, and to detect isotopic changes.

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#### $^{14}\text{C}$ BOMB PULSE IN FIRN AND ICE AT LAW DOME, ANTARCTICA

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Air bubbles in high accumulation Antarctic ice cores with unequalled time resolution are being used for the reconstruction of atmospheric concentration and isotopic records of trace gases ( $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ , etc.). Knowledge of the air trapping process which determines the age and age spread of the enclosed air is essential for these reconstructions. In this study, radiocarbon measurements have been used to characterize the trapping process.

The dry extraction technique was used to recover  $\text{CO}_2$  from ice samples taken from the DE08 and DE08/2 ice cores from Law Dome, Antarctica. The apparatus is able to process ice samples up to 2 kg in size, and the dry extraction process preserves ice crystals that may contain *in-situ* produced  $^{14}\text{C}$ , thus eliminating a possible source of interference with  $^{14}\text{CO}_2$  trapped in air bubbles. A fast recovery procedure and nearly closed system approach reduced the overall contamination in the

extraction and graphitization processes to 1.5–2  $\mu\text{g}$  of modern carbon. The *in-situ* production was found insignificant for these cores mostly due to extremely high snow accumulation rate.

Measurements are presented of the  $^{14}\text{C}$  “bomb pulse” recorded in firm air and ice air bubbles, and are compared with the expected profile derived from air diffusion studies and trapping modeling using atmospheric data as the input. High-precision measurements (up to 1% for ice samples) by AMS technique at ANSTO, Sydney, allowed the age spread of entrapped air and the air-ice age difference to be determined with high accuracy. The possibility of ice core and ice core air dating using the AMS  $^{14}\text{C}$  measurements is also discussed.

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### PROGRESS IN EXTRACTING IN SITU-PRODUCED COSMOGENIC $^{14}\text{C}$ FROM QUARTZ: IMPLICATIONS FOR INTEGRATED LATE QUATERNARY PRODUCTION RATES

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*In situ*-produced cosmogenic  $^{14}\text{C}$  (*in-situ*  $^{14}\text{C}$ ) holds great promise for estimating exposure ages and long-term average erosion rates for late Quaternary geomorphic surfaces. Reliable and reproducible *in-situ*  $^{14}\text{C}$  production rate estimates are crucial to these applications, yet they have proved very difficult to achieve. We previously reported integrated late Quaternary production rates for *in situ*  $^{14}\text{C}$  of  $19 \pm 3$  atoms  $\text{g}^{-1} \text{yr}^{-1}$ , based on whole-rock fusion analyses of one quartzite and two basalt samples collected from pristine shoreline features of well-known age and unambiguous exposure history associated with Pleistocene Lake Bonneville, Utah (Lifton *et al.* 1993, *EOS* 74: 649). Unfortunately, we have been unable to reproduce those results in subsequent analyses of the same samples.

We have thus endeavored to identify and reduce or eliminate sources of variability in our extraction procedures. Careful analysis of our results has suggested that we were unable (1) to adequately control the temperature and/or oxidizing conditions in the sample crucible using an induction furnace, and (2) to adequately remove other gases released with the  $\text{CO}_2$  during the melting of the rock. Additionally, we have found that different whole-rock samples may release their *in-situ*  $^{14}\text{C}$  at significantly different temperatures. Backstreaming oil vapors from our diffusion and roughing pumps may also have influenced some of our results to an unknown degree, while solvents and other vapors from the building vacuum lines may have affected some samples while drying in our vacuum oven.

Several steps have been taken to address these issues. We now use a resistance furnace in our extraction line, allowing more stringent temperature control, higher  $\text{O}_2$  pressures, and much cleaner operation than the previous induction furnace. Additionally, a more vigorous gas cleanup procedure has proved successful in quantitatively removing gases other than  $\text{CO}_2$  from our samples. The extraction line has been cleaned thoroughly, and we have installed equipment and instigated procedures to prevent backstreaming pump oil from entering our extraction line in the future. We have also constructed a graphitization line dedicated to preparing our *in-situ*  $\text{CO}_2$  samples for AMS analysis, with similar backstreaming protection. Additionally, our vacuum oven is now connected to a dedicated vacuum pump to eliminate the potential for contamination via the building vacuum system. Extensive testing of our blanks has confirmed that these steps have minimized past blank variability problems.

We are presently limiting our analyses to quartz separates collected from Bonneville-level, wave-cut quartzite benches to eliminate uncertainties arising from compositional and mineralogic variability. Our new procedure involves step-heating a pair of samples—one exposed at the surface, the other shielded at depth. Comparing the  $^{14}\text{C}$  thermal release patterns from each sample should allow us to identify the *in-situ*  $^{14}\text{C}$  component unambiguously. We will use these improved methods to refine our integrated late Quaternary production rate estimate for *in-situ*  $^{14}\text{C}$ . This production rate estimate should provide a strong basis for geomorphic applications of *in-situ*  $^{14}\text{C}$ , as well as providing cross-checks for production rates of other cosmogenic nuclides.

### NEUTRAL INJECTION FOR AMS

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Neutral injection was used successfully for nuclear physics experiments with helium and neon ion beams >30 yr ago and has not yet been used for AMS as negative ion beams have been found to be more convenient. However, there are many elements which form negative ions with difficulty. The positive ions of such elements can usually be generated quite easily by sputtering with negative ions. Isobar suppression can be undertaken by a variety of methods at low energies (Kilius and Litherland 1995). Neutral injection, if done by resonant electron transfer below 100 keV, gives a high quality neutral beam which can then be changed to a positive or negative ion beam in the terminal of a suitable tandem accelerator for further acceleration. The completion of the molecular destruction at high energies, and isobar separation, if necessary, has a number of advantages. Examples such as the detection of  $^{39}\text{Ar}$  and  $^{135}\text{Cs}$  will be discussed.

<sup>1</sup>Deceased January 1996

### REFERENCE

Kilius, L. R. and Litherland, A. E. 1995 Patent number 5386116, USA

### OIL FORMATION AND FLUID CONVECTION IN RAILROAD VALLEY, NV: A STUDY USING COSMOGENIC ISOTOPES AND NUMERICAL SIMULATIONS TO DETERMINE THE ONSET OF HYDROCARBON MIGRATION

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The onset of hydrocarbon migration is a critical question for the understanding of the formation process of many oil reservoirs. We present here first results for the use of cosmogenic isotopes and numerical simulations to address this question. The characteristics of  $^{129}\text{I}$  (half-life  $T_{1/2} = 15.7$  Ma; strong biophilic nature of iodine) make this isotopic system a particularly well suited candidate in this context. Although  $^{129}\text{I}$  is produced in the atmosphere by the interaction of cosmic rays with xenon isotopes, the important production mechanism for this application is that by spontaneous fission of  $^{238}\text{U}$  in the subsurface. In a non-migrating situation,  $^{129}\text{I}$  levels in hydrocarbons will reflect the secular equilibrium attained in the source rock as a function of uranium concentrations. Once migration starts, this signal will be carried along by the fluids into the reservoir rock which has in general a lower uranium concentration than the source rock. A comparison between the observed  $^{129}\text{I}$  levels in oil field brines to those supported by the uranium concentration in the reservoir rock

can be used to calculate the onset time of hydrocarbon migration. We combined this approach with numerical simulations to model the impact of crust fluid flow on oil generation within host and graben structures where the effects of convective heat transfer through fluid migration within sedimentary basin of the kind for oil formation have been recognized.

We tested this approach on samples from oil fields in the Railroad Valley, Nevada. These oil fields are found in part of the Great Basin, characterized by relatively thin crust, high thermal gradients, and high angle faulting. Commonly, these oil fields are associated with geothermal systems, most of which are active close to the center of graben structures. The simplified two-dimensional models of hydrothermal fluid and oil migration constructed for the structurally-controlled basin within two extensional faults indicated that the vertical flow rate is on the order of several millimeters per year and upwelling zones are correspondent to the oil field reservoirs. The link between structural activity, hydrothermal convection and oil maturation and migration is evident and of potential significance for the understanding of oil resources. Ages calculated using the  $^{129}\text{I}$  system in oilfield brines in Railroad Valley range from  $^6\text{Ma}$  to  $^{24}\text{Ma}$ . They indicate that brines coexisting with oil were separated from source regions at different times during the evolution of fluid migration system; the oldest age coincides with a major event of structural faulting which could have provided the heat source for the onset of geothermal convection. The oldest time estimated in our study corresponds well with the age of the initial fault movement in this area as obtained by field investigations and is in agreement with numerical model calculations for fluid convection in this area. Our results suggest that geothermal convection associated with structural activity could play an important role in hydrocarbon maturation and migration. Hydrocarbon maturation could be a result of upwelling of geothermal fluids heated by deep circulation along faults or the juxtaposition of formations with disparate temperatures as a result of faulting. Oil migration was initiated during the main episode of faulting and probably is still continuing in the Basin and Range provinces.

#### **THE TECHNICAL IMPROVEMENT IN PKUAMS**

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In the PKUAMS injection system upgrade project, we adjusted the ion source parameters to get better Cs focus and the smallest emittance. Redesigning the sample changing mechanical device provided a more reliable sample change process. The other improvement is to rebuild the injection control system by using an IPC. It is more stable, reliable and easy to use. With the improvement of sample preparation and optimized beam optics, now, PKUAMS has the ability to measure a sample that is  $<100\ \mu\text{g}$ .

#### **A DATABASE FOR SAMPLE CURATION AT PRIME LAB**

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At PRIME Lab,  $>260$  AMS samples are chemically processed, 1265 targets are measured, and 3950 unknowns, standards and blanks are loaded into sample holders per year. Every sample requires five

procedures for an AMS measurement: sample collection, chemical processing, loading into holders, AMS measurement and data analysis. Each procedure involves several pieces of data, which range from historical background to information needed to calculate results. A data management system is needed for sample tracking and for accountability. For quality assurance/quality control it is important for records to be accurate and complete.

Records containing parameters and results for all samples measured since 1993 are stored in disk files. Although these records are readily accessible (results from them are available to users on the World Wide Web), they do not contain an inclusive description of auxiliary sample data.

We are developing a full-scope sample-data management system to meet the demands for collection, chemistry, loading, measurement, evaluation, reports and billing for a large number of samples. The sample database will eliminate data redundancy, prevent data inconsistencies, link data together, allow concurrent access by several users, and maintain database integrity and security. The database will be under the control of Oracle, since it is the most widely used database management system.

### **FIRST AMS IN INDIA: PROPOSAL AND PROGRAM**

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Realizing the need for an Accelerator Mass Spectrometer (AMS) in the country, it is proposed to augment the 3 MV tandem accelerator (NEC, Pelletron, 9SDH-2) facility at the Institute of Physics, Bhubaneswar. There are two negative ion sources (SNICS and ALPHATROSS) which inject 60 keV ions into the machine through a 45 deg analyzing magnet ( $ME/Z^2 = 15$ ). There is a high resolution 90 deg analyzing magnet ( $ME/Z^2 = 151$ ,  $\Delta M/M = 0.001$  at  $\Delta X = \pm 1$  mm). The injection as well as the high energy side of the machine will soon be modified and a new beamline added to carry out measurements involving  $^{14}\text{C}$  in the first phase and  $^{10}\text{Be}$  later. The first phase of AMS applications will be in the field of Earth Sciences, especially those aspects that are relevant to the studies that are being carried out in India. These pertain to 1) global change; 2) air-sea exchange; 3) paleoseismicity; and 4) archaeology. Monsoon is the phenomenon that controls the climate and associated aspects of the Indian subcontinent and one of the ways of understanding it is through the PAGES approach of IGBP. The paleomonsoonal/paleoclimatic/paleoceanographic records are stored in the "margin" and "open" sediments of the Bay of Bengal and the Arabian Sea. These sediment cores from monsoon-induced upwelling regions of the Indian coasts are being analyzed for several "proxies" and  $^{14}\text{C}$  dating will be carried out using AMS.

There are several rivers in India which get flooded as a result of high monsoonal rainfall causing devastating effects. These waters ultimately reach the Bay of Bengal. Paleofloods, past fresh water inputs to the Bay and the paleomonsoons are all connected and a comprehensive study of these are planned; sampling is underway.  $^{14}\text{C}$  and  $^{10}\text{Be}$  measurements along with those of organic carbon and  $\delta^{18}\text{O}$  on select species of foraminifera from oceanic sector will provide an important data set of past variations and their connections for modeling. Studying paleoseismicity in the Himalayan area is another area of focus. Simultaneous efforts towards sample collection and augmentation of the Bhubaneswar machine for AMS use are underway. An update will be presented.

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## SEPARATION OF NICKEL FROM COPPER METAL FOR THE DETERMINATION OF $^{63}\text{Ni}$ USING ACCELERATOR MASS SPECTROMETRY

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Measurements of  $^{63}\text{Ni}$  ( $t_{1/2} = 100$  yr) produced by the reaction  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  could be used in the assessment of fast-neutron fluences from the Hiroshima A-bomb. Such measurements would add new information to help resolve the current discrepancy between measured thermal neutron activation values and those calculated with the DS86 dosimetry system. It has been estimated that the  $^{63}\text{Ni}$  production near the hypocenter was  $\sim 10^7$  atoms/g Cu. Because of its sensitivity, accelerator mass spectrometry (AMS) is ideal for measurements at this low level. However,  $^{63}\text{Ni}$  has to be separated from large amounts of stable atomic isobar  $^{63}\text{Cu}$  (69% of pure Cu). In this study, a procedure is presented for the electrochemical separation of small amounts of Ni from Cu. The method was developed using samples of electrical Cu wire that were irradiated with fission neutrons from a  $^{252}\text{Cf}$  source. The wire samples were electrochemically dissolved in a solution containing 1 mg of Ni carrier. The Cu was selectively deposited on a cathode at controlled potential. Measurements of total Ni after electroseparation indicate  $\sim 100\%$  carrier recovery. To prevent Cu contamination, AMS targets were prepared by nickel carbonyl generation. The AMS results show a successful quantitative separation of  $\sim 10^{-7}$  atoms of  $^{63}\text{Ni}$  from 2–20 g samples of Cu.

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## $^{10}\text{Be}$ AND $^{26}\text{Al}$ IN COSMIC SPHERULES AND ORBITAL CHARACTERISTICS OF THE INTERPLANETARY DUST

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The orbit of the interplanetary dust particles that originated in the asteroid belt, or were produced either by short period comets or high eccentric meteor streams, is gradually changed by the pressure of solar radiation. During their secular evolution towards the Sun, the dust particles are subjected to solar and galactic cosmic rays produce radionuclides such as  $^{10}\text{Be}$  and  $^{26}\text{Al}$ . A correlation between radioisotope concentrations, particle sizes and orbital characteristics can be observed even after considering the ablation process due to the atmospheric entry. For this,  $^{10}\text{Be}$  and  $^{26}\text{Al}$  concentrations in cosmic spherules of a fixed size collected from deep sea sediments were measured by means of the AMS technique. Due to the small sample weight, the carrier amount was kept very low (*ca.* 50  $\mu\text{g}$  for Be and between 100 and 200  $\mu\text{g}$  for Al). In addition, a theoretical estimation for the radioisotope distribution will be presented and compared to experimental data. This allows us to derive the orbital characteristics of interplanetary dust particles and hence their origin.

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## DETECTION OF $^{63}\text{Ni}$ AND $^{59}\text{Ni}$ BY ACCELERATOR MASS SPECTROMETRY USING CHARACTERISTIC PROJECTILE X-RAYS

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The long-lived isotopes of nickel have current and potential use in a number of applications including cosmic ray studies, biomedical tracing, characterization of low-level radioactive wastes, and neutron dosimetry. Methods are being developed at LLNL for the routine detection of these isotopes by AMS. One intended application is in Hiroshima dosimetry. The reaction  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  has been identified as one of a small number of reactions which might be used for the direct determination of the fast neutron fluence emitted by the Hiroshima bomb (Marchetti and Straume 1996, *Appl. Radiat. Isotop.* 47, 97). AMS measurement of  $^{63}\text{Ni}$  ( $t_{1/2} = 100$  yr) requires the chemical removal of  $^{63}\text{Cu}$ , which is a stable isobar of  $^{63}\text{Ni}$ . Following the electrochemical separation of Ni from gram-sized copper samples (reported by Marchetti, *et al.*, these proceedings), the Cu concentration is further lowered to  $< 2 \times 10^{-8}$  (Cu/Ni) using the reaction of Ni with carbon monoxide to form the gas  $\text{Ni}(\text{CO})_4$ . The  $\text{Ni}(\text{CO})_4$  is thermally decomposed directly in sample holders for measurement by AMS. After analysis in the AMS spectrometer, the ions are identified using characteristic projectile X-rays, allowing further rejection of remaining  $^{63}\text{Cu}$ . In a demonstration experiment,  $^{63}\text{Ni}$  was measured in Cu wires (2–20 g) which had been exposed to neutrons from a  $^{252}\text{Cf}$  source. We successfully measured  $^{63}\text{Ni}$  at levels necessary for the measurement of Cu samples exposed near the Hiroshima hypocenter. For the demonstration samples, the Cu content was chemically reduced by a factor of  $10^{12}$  with quantitative retention of  $^{63}\text{Ni}$ . Detection sensitivity ( $3\sigma$ ) was  $\sim 20$  fg  $^{63}\text{Ni}$  in 1 mg Ni carrier ( $^{63}\text{Ni}/\text{Ni} \approx 2 \times 10^{-11}$ ). Significant improvements in sensitivity are expected with planned incremental changes in the methods. Preliminary results indicate that a similar sensitivity is achievable for  $^{59}\text{Ni}$  ( $t_{1/2} = 10^5$  yr). We will also report on initial work on the application of this isotope as a biomedical tracer in living systems.

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## NORMALIZATION OF BERYLLIUM-10 IN MARINE SEDIMENTS AND ASSOCIATED PROBLEMS

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The cosmogenic radionuclide  $^{10}\text{Be}$  is predominately produced in the upper atmosphere and is quickly precipitated onto the surface of the Earth. Its production rate in the atmosphere is a function of the intensity of the geomagnetic field, solar activity, and the primary cosmic-ray flux. Thus,  $^{10}\text{Be}$  can be a valuable source of information for the previously stated geophysical and astrophysical phenomena. Nevertheless, the deposition and consequent transport of  $^{10}\text{Be}$  through the environment can obscure the original influences on its production. Presumably, such problems incurred upon transport can be limited by studying polar or mountain-ice archives because of direct precipitation of  $^{10}\text{Be}$  onto the ice. However,  $^{10}\text{Be}$  concentrations in the ice are influenced by glacial rheology and local precipitation. Potentially, marine sediments record a  $^{10}\text{Be}$  production-rate signal that is more



globally representative because of latitudinal transport and mixing of seawater. Unfortunately, interpretation of  $^{10}\text{Be}$  data measured from marine sediments is also fraught with difficulties.

Initially, it was hoped that normalization of  $^{10}\text{Be}$  to  $^9\text{Be}$  would separate the initial production-rate signal of  $^{10}\text{Be}$  from later changes in its concentration.  $^9\text{Be}$ , the stable isotope of beryllium, had been assumed to have a largely terrigenous origin. After transport to the sea,  $^9\text{Be}$  would mix evenly with  $^{10}\text{Be}$ , assuming a sufficiently long oceanic residence time. Hence, any changes in the concentration of  $^{10}\text{Be}$ , caused by depositional processes, is mimicked by the  $^9\text{Be}$  concentrations. This assumption is shown only to be partly true in our analyses of three marine sediment cores from the Gulf of California, the Sea of Japan, and the Blake Ridge in the Atlantic Ocean. The correlation of  $^{10}\text{Be}$  to  $^9\text{Be}$  varied from nonexistent at a deep site on the Blake Ridge, to relatively tight on a ridge in the sometimes restricted Sea of Japan basin.  $^9\text{Be}$ , measured from the Blake Ridge sediments, has a deep water origin separate from that of  $^{10}\text{Be}$ . In contrast, at the Sea of Japan location, both isotopes are influenced by shallow water processes.

Measurement of other elemental data is very helpful in unraveling the origins of  $^{10}\text{Be}$  in the sediments. All three sediments cores have quite different geochemical histories. Aluminum is of particular help. It clearly has a terrestrial or eolian origin in the sea and that information can be used to selectively determine the transport of beryllium. In anoxic waters such as those above and within the sediments of the Gulf of California, iron and manganese can be significantly depleted. Calcium is largely transported to the sediments by marine organisms, and dilutes  $^{10}\text{Be}$  concentrations. Magnesium concentrations, in part, are controlled by pore water chemistry in the sediments.

Of utmost importance, for the attainment of a reliable  $^{10}\text{Be}$  production-rate signal, is the sedimentation rate. The  $^{10}\text{Be}$  concentrations from the sediments from the Sea of Japan, even normalized to  $^9\text{Be}$  or the other elemental data, poorly track production-rate variations inferred from recent paleomagnetic publications. The sedimentation rate for the Sea of Japan core varied between 10 and 15 cm/kyr. A  $^{10}\text{Be}$  record consistent with the paleointensity record, for the time period *ca.* 60 to 70 kyr, was obtained from the more rapidly deposited sediments on the Blake Ridge, *ca.* 30 cm/kyr. It was necessary, though, to normalize the  $^{10}\text{Be}$  concentrations to a sum of the elemental data to remove the influences of sedimentation rate on its concentration. On the other hand, the unnormalized  $^{10}\text{Be}$  concentrations from the Gulf of California, appear to accurately track paleointensity variations. The Gulf of California sediments were largely unbioturbated and deposited at a nearly constant rate of 100 cm/kyr. Ironically, normalizing  $^{10}\text{Be}$  to the elemental data is ineffective for this core unlike the other cores, due to diagenesis of iron and manganese, infusion of aluminum in the sea during glacial meltwater events, and mixture of  $^9\text{Be}$  from variable sources. Lund *et al.* (1995) determined that a reliable paleomagnetic signal can be obtained only from those marine sediments that were deposited at rates above 30 cm/kyr. Apparently, comparable sedimentation rates are also necessary for a reliable  $^{10}\text{Be}$  record.

**COSMOGENIC  $^{10}\text{Be}$  EXPOSURE-AGE DATING OF THE POST-WISCONSIN DEGLACIATION OF INDIANA***MELANIE L. McQUINN, JON HARBOR*

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Cosmogenic  $^{10}\text{Be}$  has been measured in samples from large, well-exposed glacial erratics in Indiana, north of the Wisconsin Stage boundary. These erratics are situated on both till and outwash surfaces. The  $^{10}\text{Be}$  concentrations of five erratics from a less than one-quarter square mile area on a terrace of the Wabash River will provide information on the reproducibility of the technique, and should provide a minimum age for the terrace. In west central and northern Indiana the exposure ages of additional erratics on till surfaces should yield critical information on the deglaciation history, because they provide the only chronologic control where  $^{14}\text{C}$  data from intertill and postglacial organics are sparse or absent.

As comparisons with conventional techniques establish the accuracy and reliability of cosmogenic techniques for dating Wisconsin age tills and more recent deposits, the method will increasingly be used in efforts to refine glacial chronology by dating other erratics within the Laurentide ice sheet boundary and by correlating till units based on direct dates rather than on bracketing or absent  $^{14}\text{C}$  data. Thus, the technique has the potential to significantly improve our understanding of the chronology of the last deglaciation, by providing data in areas where other techniques are not appropriate.

**AMS TECHNIQUES APPLIED TO NEGATIVE-ION STUDIES***ROY MIDDLETON and JEFF KLEIN*

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The techniques and equipment developed for AMS studies are well suited for identifying exotic negative ions. When double-negative ions consisting of clusters of seven or more carbon atoms were suggested as the explanation for peaks of half-integral mass (molecules were assumed to be of the form  $^{12}\text{C}_{n-1}^{13}\text{C}^{2-}$ ) obtained using conventional mass spectrometry (Schauer, Williams and Compton 1990), we decided to use the AMS system at the University of Pennsylvania to incontrovertibly confirm the existence of these molecular dianions. We used our multianode  $\Delta E-E$  detector to identify the positive atomic ions produced by stripping negative-ion clusters at the terminal of the tandem. Transmission from ion source to detector was high (>10%) so that it was possible to detect many (in some cases all) of the ions produced from the breakup of a single cluster. The presence of  $2n$  positive ions of C arriving at the detector nearly simultaneously while injecting masses of  $n \cdot 12$ , provided definite proof of the existence of carbon clusters with double-negative charges. To examine the physical structure of the dianions, we replaced several stripper foils at the terminal of the tandem with track detectors consisting of very thin ( $\sim 2 \mu\text{g}/\text{cm}^2$ ) carbon foils positioned  $\sim 1$  mm from Lexan sheets. The Coulomb explosion in the carbon foil resulted in separations of several tens of micrometers between the tracks produced by the ionized atoms from the breakup of a single molecule. Count rates were kept low enough so that the clusters of tracks from individual molecules were distinct. Similar searches for polyatomic dianions of Li, B, O, and Si were unsuccessful.

We have also studied the polyatomic anions (singly charged) of several light elements of general interest to the AMS community. Somewhat surprisingly, we have detected currents of stable  $^{14}\text{N}_2^-$ , and  $^{14}\text{N}_3^-$  ions with lifetime  $>10$  ms. Use of AMS techniques allowed us to distinguish between diatomic N (mass 28, 29, or 30) and monatomic Si, and between triatomic molecules of N and other possible molecules with the same mass made of other elements.

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#### MODIFICATIONS TO PRIME LAB CESIUM SPUTTER ION SOURCE

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The spherical-ionizer cesium sputter ion source used at PRIME Lab was purchased from Southern Cross Corporation (Model IS-1A). This source is now being sold by High Voltage Engineering Europa B.V. (HVEE). Our source is identical to those used at the Woods Hole Oceanographic Institution radiocarbon facility except it contained a single-sample changer. A multi-sample changer has been constructed to increase the sample throughput. The new freon-cooled rotating copper wheel holds up to eight sample cathodes and can be rotated in either the clockwise or counterclockwise directions. A precision indexing Geneva drive rotates the wheel, whose axis is parallel to but displaced from the source axis. Four opto-reflective sensors measure the sample position and relay the information through a fiber-optic system to the computer CAMAC crate. The sample position is changed by the computer in about three seconds. Through the use of a dual shaft rail system and a vacuum lock a wheel can be changed in a few minutes using a semi-automated vacuum pumpdown system.

We have made several improvements to the ion source. We redesigned the cesium reservoir and feedtube assembly to incorporate a conflat flange design and eliminate the outer feedtube heater jacket. We found that large stable chlorine beam currents require good ion source pumping to avoid memory effects, so the source vacuum was improved by enlarging all pumpout holes where possible. A new outer shield was installed to improve the cesium shielding of the cathode insulator. Finally, an improved ionizer shield was constructed with eight uniformly spaced spray holes for a more uniform cesium spray. These improvements have resulted in typical AMS operating currents of  $5\ \mu\text{A}$  for BeO,  $300\ \mu\text{A}$  for Al,  $3\ \mu\text{A}$  for I, and  $8\ \mu\text{A}$  for Cl.

Chlorine-36 measurements require the suppression of the stable isobar  $^{36}\text{S}$ . We mount our AgCl samples on a surface of reagent-grade AgBr, which fills a 8 mm diameter hole drilled into our 9.5 mm diameter copper cathodes. Negative ion formation is suppressed on the AgCl and AgBr surfaces. The ratio of the rate of  $^{36}\text{S}^-$  to  $\text{Cl}^-$  ions leaving the source ranges from 10 to  $1000 \times 10^{-12}$  depending on ion source conditions, sample size, and quality of the chemical sample preparation.

**<sup>36</sup>Cl - A POTENTIAL DATING TOOL**

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The need for an isotopic tool capable of extending our ability to date fossil organic materials beyond the 50,000-yr range of <sup>14</sup>C has been recognized for some time. Chlorine-36, a cosmogenic nuclide with a half-life of 300,000 yr, appears to be eminently suited to this role. It exchanges readily with stable chloride, which is present in ppm quantities in most living materials, and can be measured accurately at extremely low levels by accelerator mass spectrometry.

Dating is most commonly performed on fossil remains of organic material, some of the more common being wood, pollen, seeds, peats, and small organic lenses located in strata composed of mainly inorganic detritus. Largely inorganic materials such as bones and teeth are also sometimes suitable for dating. In order to demonstrate the applicability of <sup>36</sup>Cl to paleodating, it is necessary to prove both the maintenance of a unique signal for this isotope in any given growing season, and the retention of that signal during burial and diagenesis.

Using AMS techniques to measure <sup>36</sup>Cl/Cl, we have shown that good agreement exists between this ratio in plant leaves and seeds and that in the soil pore water utilized by the plant, suggestive of long-term equilibrium between seed and precipitation values. We subsequently identified the large pulse of <sup>36</sup>Cl injected into the stratosphere during atmospheric weapons testing, recorded in conifer seeds grown in the 1950s, and hence providing verification of the uniqueness of the annual input signal.

In order to prove that this signal is retained over long time frames, we have made some preliminary measurements of <sup>36</sup>Cl and stable chloride in peats and organic-rich sediments. The non water-leachable chloride component of a "modern" peat (<sup>14</sup>C "age"? 6000 yr BP) has a <sup>36</sup>Cl/Cl component similar to those we have measured in pre-1950 seeds and other vegetative materials grown early in this century. <sup>36</sup>Cl/Cl measurements in older organic-rich sediments previously dated by optical dating methods, and some measurements in mastodon teeth, are in progress.

**A NEW INTERPRETATION OF THE DISTRIBUTION OF BOMB-PRODUCED  
CHLORINE-36 IN THE ENVIRONMENT, WITH SPECIAL REFERENCE TO THE  
LAURENTIAN GREAT LAKES**

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In the early days of AMS, <sup>36</sup>Cl was thought to be an ideal hydrological tracer. For old waters, except in rare cases, *e.g.*, the Great Artesian Basin in Australia (Bentley *et al.* 1986), this hope proved vain. The difficulties were proved to be the result of <sup>36</sup>Cl production by other than cosmological means—in particular, production in surface rocks by cosmic ray neutrons, and production by fission neutrons in the aquifer. Nevertheless, it was still believed that chlorine in the hydrological environment existed primarily in the ionic form, Cl<sup>-</sup>, and was "conservative", that is, it moved with the water at the same rate, and hence for waters younger than *ca.* 50 yr, the bomb-produced pulse of <sup>36</sup>Cl would still be useful as a tracer. However, vastly improved information on Cl behavior in vegetation, litter and surface soils (Milton *et al.* 1995) brings the "conservative" assumption into question. Strong

support for this new interpretation comes from the previous knowledge that Cl in the atmosphere is found largely in organic compounds (Rahn *et al.* 1976).

It is now clear that Cl is recycled by vegetation, and furthermore is held up for considerable periods in the ground waters of at least some of the seasonal wetlands that cover a large fraction of the Canadian Shield, and in particular, the drainage basin of Lake Superior. This new knowledge is used to improve our understanding of the concentrations, previously considered anomalous, of  $^{36}\text{Cl}$  in the Great Lakes (Milton *et al.* 1994), and may be able to throw some light on the very large variation in the measured amounts of bomb-produced  $^{36}\text{Cl}$  at different geographic locations. It is also pertinent to the movement of the many chlorine-containing pollutants in the biosphere.

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## COSMOGENIC RADIONUCLIDE CONTENTS OF ANTARCTIC METEORITES FROM ALLAN HILLS HAVING HIGH NATURAL THERMOLUMINESCENCE

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We analyzed a group of 15 H<sub>4</sub>-H<sub>6</sub> chondrites recovered from the Main Allan Hills blue ice field of Antarctica. Previous studies indicate these chondrites exhibit high TL level (>100 krad) unusual by most meteorite standards (Benoit and Sears 1993).  $^{14}\text{C}$  analysis of two meteorites reveal terrestrial ages  $\leq 2300$  yr (Benoit and Sears 1993). At Purdue's PRIME lab facility, we determined  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{36}\text{Cl}$  activities that range from  $15.4 \pm 0.40$  to  $18.4 \pm 0.35$  dpm kg<sup>-1</sup> meteorite,  $38.1 \pm 1.18$  to  $58.0 \pm 2.14$  dpm kg<sup>-1</sup> meteorite and  $18.4 \pm 0.61$  to  $23.9 \pm 0.60$  dpm kg<sup>-1</sup> metal, respectively. An unusual three-isotope plot of  $^{10}\text{Be}/^{26}\text{Al}$  vs.  $^{36}\text{Cl}/^{26}\text{Al}$  illustrates a highly significant positive linear correlation with a 0.906 coefficient, corresponding to a p-value  $\ll 0.001$ . These results demonstrate a relationship between natural TL and cosmogenic isotope abundance for at least one suite of samples. These two parameters may be simultaneously used to determine the irradiation and orbital history these meteorites have experienced (Benoit and Sears 1991).

The high TL correlation differs markedly from that involving Antarctic meteorites previously analyzed at Purdue (Michlovich 1994). Sampling plays an important role when comparing these two populations. While the earlier sampling involved 43 H-chondrites for which volatile trace elements had been determined by RNAA, our sample selection was based strictly on TL levels. Previously analyzed H-chondrites were randomly selected to minimize both pairing and weathering for RNAA purposes. On a three-isotope plot, prior data yield a correlation coefficient of 0.388, only a weak correlation. These results require a correction due to long terrestrial ages, increasing the correlation coefficient to 0.544, still significantly lower than that of the high TL suite.

Despite differences in petrographic type, H4-6, the high TL meteorites may derive from a single parent object as suggested by Benoit and Sears (1993). Radionuclide activities are all close to saturation values, indicating that the parent meteoroid was relatively small. The three-isotope plot for high TL samples suggests a simple irradiation history, which also supports the notion of a relatively small sized object. In addition to saturated radionuclide concentrations, the unusually high TL levels are consistent with the suggestion that the meteorite suite experienced an orbital change from  $\geq 1.1$  A.U. to 1.0 A.U. within the last 105 yr: samples were irradiated at perihelia  $\geq 1.1$  A.U. and were unable to adjust TL levels to the warmer temperatures at 1.0 A.U. before entering the Earth's atmosphere. By comparing radionuclide concentrations, TL data, petrographic types, geographic locations, and terrestrial ages, we conclude that these meteorites are fragments of a single parent object that experienced an unusual orbital change over a short time period ( $< 105$  yr). Currently, AMS studies are underway to determine the  $^{14}\text{C}$  content of each meteorite.

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#### SURFACE $^{129}\text{I}/^{127}\text{I}$ RATIOS – MARINE VS. TERRESTRIAL

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$^{129}\text{I}/^{127}\text{I}$  ratios measured in surface marine materials such as seawater, recent marine sediments, and modern seaweeds, are compared with ratios measured in surface terrestrial materials such as freshwaters, soils and terrestrial plants. At the Earth's surface the  $^{129}\text{I}$  inventory is dominated by anthropogenic inputs. Away from point sources (nuclear fuel reprocessing facilities), anthropogenic  $^{129}\text{I}/^{127}\text{I}$  ratios measured in terrestrial materials are at least an order of magnitude greater than those from marine materials. For example, surface seawater from the Gulf of Mexico has a ratio of approximately  $60 \times 10^{-12}$ , while ratios measured in freshwater from the Trinity River on the adjacent continent are *ca.*  $1100 \times 10^{-12}$ . Likewise, terrestrial and aquatic plants exhibit higher ratios than seaweeds (on the order of  $10^{-8}$  and  $10^{-10}$ , respectively). Ratios from surface soils and marine sediments near the sediment/water interface show a similar relationship. This is perhaps not surprising, given that ambient stable iodine concentrations are in general at least an order of magnitude lower for terrestrial materials than for their marine counterparts.

Data from soils and plants from Texas and from Western New York, along with an estimate for the current atmospheric ratio from rainwater and epiphyte samples will be presented. High ratios in rivers and lakes apparently reflect bomb fallout plus a component of atmospherically-delivered fuel reprocessing deposition. This suggests that this iodine has a relatively short residence time in watershed soils and plants. Fundamental unknowns are the residence time of various forms of iodine in soils, the stability of  $^{129}\text{I}/^{127}\text{I}$  ratios in soils and biota, and the nature and level of the atmospheric anthropogenic component.

The large difference between marine and terrestrial  $^{129}\text{I}/^{127}\text{I}$  ratios has several potential uses. We have begun a project for which the goal is to establish  $^{129}\text{I}$  as a tool to identify and trace terrestrial organic carbon in marine sediments. Post-bomb organic material in sediments from coastal areas could be characterized as primarily derived from terrestrial or marine inputs, based on the  $^{129}\text{I}/^{127}\text{I}$  ratios measured. This information is crucial in global carbon cycle models, since the largest marine

reservoir for carbon is continental shelf sediments. In addition, anthropogenic  $^{129}\text{I}/^{127}\text{I}$  ratios can be used to study the biogeochemical iodine cycle, about which little rate-related information is available. Thus,  $^{129}\text{I}$  could be used as a surrogate for studying the movement of the short-lived biohazard  $^{131}\text{I}$  through the biosphere, and for the study of releases and transport of iodine radionuclides from nuclear facilities.

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## PREPARATION OF ANTHROPOGENIC $^{129}\text{I}$ IODINE SAMPLES USING CARRIER IODINE - POTENTIAL PROBLEMS

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The requirement of a 1-mg iodine sample for  $^{129}\text{I}/^{127}\text{I}$  ratio measurement by AMS often presents a significant challenge in the chemical preparation of these samples. Post-bomb materials generally have high enough ratios that carrier iodine of known ratio is added to increase the bulk of the final silver iodide sample. Virtually all anthropogenic ratios reported in the literature are calculated from a weighted average, using measured values for the weight of iodine from the sample and carrier, the ratio of the carrier, and the ratio of the combined sample.

We performed two laboratory tests to determine the accuracy of the carrier method for post-bomb  $^{129}\text{I}$  seawater and seaweed samples. A repeated extraction was performed on a 1-L surface seawater sample, with 2.5 mg of I carrier (as iodate) added at the beginning of the procedure. The seawater + carrier solution was left overnight, then iodine was extracted into carbon tetrachloride, and back-extracted into a sulfuric acid/sodium bisulfite solution, and the iodide in solution was precipitated using silver nitrate. The procedure was repeated two more times on the "waste" (+ additional carrier iodine) from the  $\text{CCl}_4$  extraction. Results show that the measured ratio approaches the ratio of the carrier slowly, suggesting that the iodine in the seawater matrix is extracted at <100% efficiency into the  $\text{CCl}_4$ . The ratio of carrier to seawater iodine (which in the first case is 5:1) may also effect the extraction efficiency. The third extraction had a ratio that was within  $2\sigma$  of the measured ratio for the carrier I.

We also extracted iodine from modern seaweeds using the alkali leach and fusion method, adding carrier at different stages of the extraction, and as a control, making samples from the iodine-rich *Laminaria* without carrier. Two mg of carrier I (as iodide) was added, in one experiment as the leachate became liquid during heating, and in a second, after back-extraction into the sulfuric acid/sodium bisulfite solution. In both cases, the calculated ratio was higher by a factor of two to three than the ratio measured on the sample without carrier. Agreement was better in the case in which carrier was added at the later stage. It appears that carrier I may be lost during sample preparation, in greater proportion than organically bound I from the seaweed.

Two potential pitfalls are evident. One is that even if the carrier iodine is in the same oxidation state as iodine from the sample, extraction efficiencies for the carrier and sample iodine may differ due to matrix effects. Another is that accurate measurement of stable iodine concentrations at various stages during the extraction is imperative. Given that determination of iodine at low concentrations, in solids, and in matrices with high dissolved solids is not at all trivial, the absolute accuracy of some reported anthropogenic ratios may be in question.

### **INVESTIGATION OF CHANGING BIOSPHERE BY $^{14}\text{C}$ -DATING OF PEAT PROFILES FROM BAVARIA**

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As part of the program "Change of the Geo- and Biosphere During the Last 15,000 Years" some peat sediment profiles from South Bavaria, provided by H. J. Küster, have been dated using the Erlangen AMS facility (Arslan *et al.* 1994; Kretschmer *et al.* 1996). The sediment cores have been roughly predated by pollen-analytical methods, but due to local climatic variations this method has an estimated error of up to 2000 yr. With several sediment datings together with pollen analyses it should be possible to establish a better chronology of climate and vegetation of Holocene in Bavaria. By our measurements we found that the Eggstätt peat with an age of 13,000 BC is one of the oldest continuous peats in South Bavaria.

The sediment samples were pretreated with the usual acid-alkali-acid method to remove carbonates and humic acids. The remaining material was converted to  $\text{CO}_2$  by heating it up to  $900^\circ\text{C}$  with  $\text{CuO}$  and silver wool under vacuum. Then the  $\text{CO}_2$  was catalytically reduced with  $\text{H}_2$  and iron powder at a temperature of  $625^\circ\text{C}$ . These so called bulk sediment samples were dated by AMS.

To eliminate dating errors due to the "hard water effect" we try to date extracted pollen itself. The first step of the preparation procedure is the treatment of the sample with hot  $\text{NaOH}$  to remove humic acids. Afterwards the sample is sieved with a  $100\ \mu\text{m}$  nylon mesh. The filtrate is then repeatedly heated with  $\text{NaOH}$  followed by  $\text{HCl}$  to remove carbonates. As described in Faegri and Iversen (1989) we use for the removal of silicates hot  $\text{HF}$  and  $\text{HCl}$ , for the deflocculation of amorphous organic material a treatment with  $\text{NaOCl}$  is performed, and at last the cellulose of the residues is removed with  $\text{H}_2\text{SO}_4$ . After each step the dissolved material is separated from the pollen by sieving with a  $20\ \mu\text{m}$  nylon mesh. The efficiency of this separation method is finally checked by microscope. The adjacent conversion to sputter targets is done in the usual manner described above. But due to the small sample size the reduction is performed in a newly built reduction facility with a volume of  $3.8\ \text{cm}^3$ . The resulting material contains  $50\text{--}500\ \mu\text{g}$  carbon. First pollen datings have been performed and the results are encouraging. Future measurements with pollen will lead to a more reliable dating of sediments.

This work was supported by the Deutsche Forschungsgemeinschaft.



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## **A COMPACT $^{14}\text{C}$ ISOTOPE RATIO MASS SPECTROMETER FOR BIOMEDICAL APPLICATIONS**

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The absence of molecular interferences and isobar suppression has given AMS its superior sensitivity in the field of isotope ratio mass spectrometry and has made the detection of long-lived radio-isotopes possible. In the case of  $^{14}\text{C}$ , AMS rejects the isobar  $^{14}\text{N}$  completely, as nitrogen does not form stable negative ions which are needed for injection into tandem accelerators. Consequently,  $^{14}\text{C}$  can be easily detected with small tandem accelerators and terminal voltages as low as 2–3 MV. Moreover, it has been demonstrated that  $^{14}\text{C}$  can be detected at a terminal voltage of only 1.4 MV, using charge state  $2^+$  (Lee 1992).

In recent years, interest has increased in applying  $^{14}\text{C}$  in biomedical studies. Here, the high sensitivity of AMS allows the study of the kinetics and the pathway of toxins, mutagens and carcinogens at levels corresponding to actual environmental exposures (Davis 1994).

In this article we present the performance and layout of a small AMS  $^{14}\text{C}$  systems especially designed to the requirements of biomedical research.

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## **STATUS OF THE PRIME LAB UPGRADE**

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The first phase of the upgrade program currently underway at PRIME Lab has been completed. This phase of the upgrade included the installation of an NEC Pelletron charging system, shielded col-

umn grading resistors, spirally-inclined titanium-electrode acceleration tubes, an 8-position sample changer for the ion source, electrical filtering of all low voltage wiring entering the accelerator pressure vessel, new software for controlling the injector and analyzing magnets, new cryopumps, and the removal of much obsolete equipment. Operational experience with the initial phase of the upgrade demonstrates great improvement in the useful operating voltage, the frequency of tank sparks which were reduced by more than an order of magnitude, the beam transmission, and the beam stability. This and other improvements have resulted in a factor of two improvement of the AMS precision for  $^{36}\text{Cl}$  measurements and a factor of 3 to 5 improvement in sample measuring capacity.

The next phase of the upgrade is currently underway. This phase will include rebuilding the beam line between the source and the injector magnet, and rebuilding the stripper assembly in the accelerator terminal, and the integration of the controls for this equipment into the VISTA control system which will ultimately control the entire accelerator.

Most of the components for the beam line have already been purchased, and a large number of custom components have been sent to the machine shop for construction. It is expected that most of the components will be finished by the time of the AMS7 conference. Most of the drawings for the terminal components are completed. A large pressure test vessel (a used small accelerator tank) has been purchased and is currently being installed. The entire terminal stripper assembly will fit in this test fixture to permit pressure testing and leak checking of most of the components before the accelerator is shut down for this part of the upgrade. Several critical components for the terminal, such as one of the turbo pumps, are ready for pressure testing. The upgrade of the terminal will include the installation of a fiber-optic communication system to permit computer control of the new terminal components. These improvements are expected to make a substantial improvement in the beam transmission and stability for AMS measurements.

Initial planning is underway for the installation of the new AMS injector. Only minor changes have been made to the design presented in AMS6. This design will provide for the ability to do fast switching once the positive ion analysis portion of the accelerator is also upgraded. The computer control system will be integrated into the accelerator systems during the upgrade to eliminate the need for the construction of manual controls.

#### **THE LONG-TERM VARIATION OF $^{10}\text{Be}$ FLUX CHANGES AT ODP SITE $^{925}\text{B}$ ON THE SEARA RISE IN THE EQUATORIAL ATLANTIC**

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In recent study, the radionuclide  $^{10}\text{Be}$  (half life:  $1.5 \times 10^6$  yr) has been used in flux studies as a geochemical and a sedimentological tracer. The progress of flux studies has provided opportunities to understand its behavior, its residence times in the ocean and the process by which it is removed to the sediments (*i.e.*, boundary scavenging).

Our primary goal in this study was to assess the long-term variation of  $^{10}\text{Be}$  flux changes and to estimate the source of fluxes on the Seara Rise in the equatorial Atlantic.

Age-corrected  $^{10}\text{Be}$  flux concentrations range from 40 to  $140 \times 10^7$  atoms/cm<sup>2</sup> (average  $60 \times 10^7$  atoms/cm<sup>2</sup>). The variations of  $^{10}\text{Be}$  flux can be explained by source change in the sediments. We can mention at least the following conclusions from the present study.

1. The highest values of  $^{10}\text{Be}$  flux after 1 Ma reflect remarkably an increased lateral terrigenous input from the Amazon drainage area.
2. The higher values of  $^{10}\text{Be}$  flux before 6 Ma were influenced by an enhanced vertical input causing the high paleoproductivity in the ocean surface.
3. The lower values of  $^{10}\text{Be}$  flux near *ca.* 4 to 5 Ma might be explained by low paleoproductivity in the ocean surface and/or winnowing effects with bottom current, although a precise determination of the cause could not be obtained.

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## $^{14}\text{C}$ AMS MEASUREMENTS ON N- AND S-HEMISPHERE TROPICAL TREES IN THE BOMB-PULSE PERIOD

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Tropical and temperate forests have considerable relevance in the global ecosystem and are a significant component of the biosphere and carbon cycle. It is considered that they also have an important role in damping the  $\text{CO}_2$  content of the atmosphere, which is currently increasing at a constant rate of *ca.* 0.5% per year. Growth rate information about tropical and temperate trees, especially species with commercial value, is also necessary to allow management and planning for planting, harvesting and rotation times. Growth rate estimates can also be used to calculate biomass gains/losses.

Unlike trees of temperate latitudes, most tropical tree species do not exhibit annual growth rings. Consequently, information from a major, climatically important region is unavailable for modeling and does not contribute to dendroclimatology.

However, teak (*Tectona grandis*) is a tropical tree species which does exhibit annual growth rings. This arises because of the deciduous nature of teak, and not from significant temperature ranges over the seasons. However, year-to-year ring width variation is governed by the level of precipitation and the season. We report radiocarbon measurements using the ANTARES AMS spectrometer from the bomb-pulse period 1955–1980 on cross-dateable teak tree samples from both the Northern (India) and Southern hemispheres (Java). Based on six samples over the 25-yr period 1955–1980 we find the measured  $\Delta^{14}\text{C}$  values to agree closely with atmospheric  $^{14}\text{C}$  concentrations from the appropriate hemisphere.

**A NEGATIVE ION SURVEY***M.-J. NADEAU,<sup>1</sup> A. E. LITHERLAND,<sup>2</sup> X.-L. ZHAO,<sup>2</sup> M. A. GARWAN<sup>3</sup> and L. R. KILIUS<sup>2,4</sup>*

The negative ion properties of many of the elements of the periodic table have been known for some years; however, there are some 31 elements for which conclusive experimental evidence for the existence of a stable negative ion has not been found. After a brief overview of the experimental method used, we will present the results obtained for the three elemental groups studied (group II, lanthanides, actinides). We will then conclude stressing the similarities between different elements and different elemental groups. We will also show the evolution in our knowledge of negative ions since our last presentation at this conference.

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The new facility, accepted in September 1995 from High Voltage Engineering Europa, is similar in design to the NOSAMS Facility at the Woods Hole Oceanographic Institution and the AMS system of the University of Groningen, all based on a 3MV tandem accelerator and the simultaneous injection of the three carbon isotopes. Three aspects of the system will be addressed: the stability of the system, the background inherent to the spectrometer and the response of the isotope ratios to variations of different parameters of the system. The system has shown very encouraging performances during the acceptance tests and other stability tests performed afterward; the result of these will be presented and compared with the result of a sequential measurement (by opposition to simultaneous injection). We will also discuss the different sources of spectrometer background and the steps taken to reduce it. The system has also shown to be quite insensitive to small variations such magnetic and electrostatic drifts. The effect of such variations on the isotope ratios in comparison with the accuracy and stability of the respective power supplies will be presented. The issue of the symmetry of the recombinator relative to the three carbon beams will also be mentioned.

<sup>1</sup>Leibniz-Labor für Altersbestimmung und Isotopenforschung, Christian-Albrechts Universität, Kiel, Germany<sup>2</sup>High Voltage Engineering Europa B.V., Amersfoort, The Netherlands**LATITUDINAL DISTRIBUTIONS OF <sup>7</sup>Be AND <sup>10</sup>Be IN THE ATMOSPHERE***H. NAGAI,<sup>1</sup> M. UCHIDA,<sup>1</sup> M. MURAYAMA,<sup>2</sup> N. AHAGON,<sup>3</sup> M. IKEHARA,<sup>3</sup> M. IMAMURA,<sup>4</sup> S. HATORI<sup>5</sup> and K. KOBAYASHI<sup>5</sup>*

Distributions of cosmogenic <sup>7</sup>Be and <sup>10</sup>Be in the atmosphere were measured for the area between latitudes of 60°S–40°N and longitudes of 140–170°E over the Pacific Ocean and the Antarctic Sea. Samples were collected during two expeditions of R/V “Hakuho-Marū”, KH94-3 (September–October 1994) and KH94-4 (December 1994–February 1995). The samples of atmospheric particulate were collected on filter papers at a flow rate of 1 m<sup>3</sup>/min for *ca.* 24 h using a high-volume air sampler. Activities of <sup>7</sup>Be in the filter papers were directly measured by HP-Ge detectors within 2 months after sample collection. For measurements of <sup>10</sup>Be, each half of the filter papers was ashed

and treated with 2N HCl. After adding 0.1–0.2 mg  $^9\text{Be}$  carrier,  $^{10}\text{Be}$  was purified by cation exchange column. AMS measurements were performed with the MALT 5UD Pelletron at the University of Tokyo.

Observed concentrations of  $^7\text{Be}$  in the atmosphere ranged from  $10^3$  to  $5 \times 10^4$  atoms/m<sup>3</sup>, while  $^{10}\text{Be}$  ranged from  $10^4$  to  $10^5$  atoms/m<sup>3</sup>. Although the concentrations of these two nuclides largely varied, similar latitudinal distributions were found. The distributions have two maxima at 20–30° in both hemispheres and two minima at the equator and 60°S. The concentration ratios  $^{10}\text{Be}$  to  $^7\text{Be}$  were *ca.* 1.3 in the northern hemisphere but increased in the southern hemisphere by factor 2 to 3. This high  $^{10}\text{Be}/^7\text{Be}$  value may be attributed to the larger air flow from stratosphere to troposphere in the southern hemisphere than in the northern hemisphere, or to the increase of the production rate ratio  $^{10}\text{Be}$  to  $^7\text{Be}$  with decreasing cut-off rigidity.

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## ACCELERATOR MASS SPECTROMETRY AT THE KYOTO UNIVERSITY TANDEM ACCELERATOR

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The Kyoto University 5MV tandem Van de Graaff was replaced by an 8UDH pelletron in 1990 (Nakamura *et al.* 1988). This machine is mainly used for nuclear science, interdisciplinary field using ion beam and education of students for nuclear and its related science.

The AMS project is being developed to determine long-lived radioisotopes, *e.g.*,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$  and  $^{129}\text{I}$  and also  $^{14}\text{C}$  for checking the whole AMS system. A high intensity 60-sample ion source (High Voltage Engineering Europa) is placed on a 200-kV injector deck. The low energy beam line includes an off-axis Faraday cup for measuring a main isotope beam (*e.g.*,  $^{14}\text{C}^-$  ion beam) and a mesh system for beam attenuation. In the high energy section, the 90 deg analyzing magnet has been fitted with additional small pole tips to allow good transmission of all isotopes of interest. An off-axis slit system is used for stabilizing the terminal voltage using an ion beam produced by a molecular beam, *e.g.*, a  $^{13}\text{C}_4^+$  beam produced by  $^{13}\text{CH}^-$  beam.  $^{14}\text{C}_4^+$  ions were separated from  $^{14}\text{N}_4^+$  and  $(^7\text{Li}_2)_4^+$  ions by use of thin Al foil (3 microns) and a silicon energy detector.

The experience at the Kyoto University tandem accelerator AMS system will be discussed.

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**AMS <sup>14</sup>C CHRONOLOGICAL STUDY ON HOLOCENE ACTIVITIES OF ACTIVE FAULTS IN JAPAN**

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Slipping of active faults generates earthquakes. If we can detect traces of ancient slips of active faults, we can correlate them with earthquakes that occurred in the past. The scale of a slip correlates with the time interval between the slip and the previous one during which strain accumulated in the fault system. Thus it is very important to examine the history of fault activities in the late Quaternary, to provide a risk estimation of the next slip time for prevention of disasters. To establish chronology of past slips of active faults, AMS <sup>14</sup>C dating of macro fossil, charcoal and soil organic material samples collected around the fault is very promising, because the method requires carbon samples of very small size.

Some active faults were discovered in a shallow sea area. They provided us an ideal field to study behavior of a fault during the Holocene, because sea sediments deposited continuously, by recording seismic events of the faults in them. The following procedures are applied to two submarine active fault systems located in the northwestern part of Beppu Bay, eastern Kyushu and off the coast of the town of Futami in the northwestern Shikoku, Japan: 1) surveying an exact line of a submarine fault and continuation of sediment layers on both sides of it by using a high-resolution single-channel seismic profiler; 2) collecting sediment samples on both sides of the fault by using a piston core sampler; 3) analyzing core samples for magnetic susceptibility, micropaleontology and lithology; 4) correlating exactly each sediment layer of the cores on both sides of the fault to recognize several seismic events by relative vertical displacements of relevant layers between the two core sediments; 5) establishing chronology of the events by AMS <sup>14</sup>C dating of mainly shell fossil samples selected from the sediments.

We also applied AMS <sup>14</sup>C dating to establish the chronology of the Nojima fault, located in Awajishima, Hyogo Prefecture, which is one of the faults that generated the 1995 Hyogoken Nanbu Earthquake that occurred on 17 January 1995 and destroyed the city of Kobe. Preliminary chronological results are: 1) the Nojima fault has repeated the displacements a few times in the last 2000 yr; 2) a slip definitely occurred along the Nojima fault *ca.* 2000 yr ago; 3) several types of deformation, including an open crack, were likely to be formed close to the fault, associated with an earthquake that occurred 400 yr ago, that is probably assigned as the 1596 Keicho Earthquake.

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**ENVIRONMENTAL <sup>90</sup>Sr MEASUREMENTS**

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<sup>90</sup>Sr (T=28.5 yr) is a long-lived anthropogenic radionuclide produced in fission. The current and historical distribution of <sup>90</sup>Sr in the environment is useful for the prediction of the behavior of this nuclide released into the environment. The rapid radiochemical detection of <sup>90</sup>Sr in environmental

samples poses non-trivial problems mainly because its pure beta activity is masked by a shorter-lived strontium isotope  $^{89}\text{Sr}$ , also a fission product. AMS offers a direct, fast and sensitive measurement of  $^{90}\text{Sr}$  concentration by atom counting, the major detection problem being in this case the interference of stable isobaric  $^{90}\text{Zr}$  impurities. The  $^{90}\text{Y}$  isobaric daughter reaches secular equilibrium where  $^{90}\text{Y}/^{90}\text{Sr} = 2 \times 10^{-4}$  and is insignificant in atom counting. AMS measurements of  $^{90}\text{Sr}$  were performed with the Rehovot 14UD Pelletron accelerator at a terminal voltage of 11 MV using our standard detection system. Samples of  $\text{SrH}_2$  were prepared by reduction of  $\text{SrO}$  to  $\text{Sr}$  and conversion to hydride in a hydrogen chamber at 400 deg C.  $\text{SrH}_3^-$  are the preferred ions owing to high beam intensity and low Coulomb explosion effects. The ions were identified by measuring time-of-flight, total energy and three independent energy loss signals in an ionization chamber filled with 15.7 torr of isobutane. The measurements are normalized to a known standard of  $^{90}\text{Sr}/\text{Sr} = 4.3 \times 10^{-11}$ . For this standard 400  $^{90}\text{Sr}^{11+}$  ions were counted in 13 min. A ratio of  $^{90}\text{Sr}/\text{Sr} = 2 \times 10^{-13}$  was measured for a blank sample (3 counts in 33 min), probably due to the vicinity of the intense group of  $^{90}\text{Zr}$  ions in the spectra. This sensitivity corresponds in practice to a  $^{90}\text{Sr}$  specific activity of 0.2 pCi/L in environmental samples, a factor of *ca.* 5 better than typically achieved in standard beta counting methods. Environmental samples are being prepared and studied with our AMS system.

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## EXPANDED SMALL SAMPLE CAPABILITIES AT NOSAMS: PREPARATION AND ANALYSIS OF CONVENTIONAL GRAPHITE TARGETS CONTAINING $\leq 0.15 \mu\text{g}$ CARBON

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Eighty small AMS samples containing between 15 and 150  $\mu\text{g}$  carbon were prepared by reduction of  $\text{CO}_2$  over cobalt metal at  $625 \pm 25^\circ\text{C}$  in the presence of excess  $\text{H}_2$ . Analysis of the graphitization procedure shows that optimal reduction conditions favor temperatures below  $608^\circ\text{C}$ , and success rates improve with larger sample sizes. AMS results for the 41 OxI and OxII standards found that 84% of targets containing  $\geq 20 \mu\text{g}$  carbon had "good" (accurate to within 2% of "true" and imprecision of  $\sim 2\%$ ) performance, while only 20% of targets containing  $< 20 \mu\text{g}$  carbon were classified "good". Mean fm results are  $1.051 \pm 0.015$  (HOxI) and  $1.364 \pm 0.015$  (HOxII), for an overall imprecision of  $\pm 15\%$ . The graphitization blank is insignificant—equivalent to  $\sim 0.01$  mmol modern carbon.

Detailed analysis of the raw AMS data suggests that machine efficiency issues are critical below the 100  $\mu\text{g}$  threshold and must be addressed to achieve high quality results on small samples. Sample  $^{12}\text{C}$  current output (I12) scales linearly with sample size. The low currents associated with small samples in turn affect the isotopic ratios  $^{14}\text{C}/^{12}\text{C}$  and  $\delta^{13}\text{C}$ . The depression in  $^{14}\text{C}/^{12}\text{C}$  ratio is especially significant and is not believed to be the result of an unquantified "dead" carbon blank. While application of a size-calibrated model correction factor to all calculations is possible, the use of size-matching standards is recommended as the preferable option for analysis of sub-milligram samples.

**REDUCING THE MAGNET SWITCHING TIME FOR AMS AT PRIME LAB***M. PERRY, D. ELMORE, F. RICKEY*

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Although we have plans for a fast-switching system (“Status of PRIME Lab Upgrade”, K. A. Mueller *et al.*, abstract presented at this conference), for the first few years of operation at the Purdue Rare Isotope Measurement Laboratory we are cycling magnets to obtain isotope ratios. To increase the precision, it is important to minimize the total cycle time. The magnet change time is a significant fraction of the total cycle time since the magnets must be steady to within a few hundred ppm before measurements can begin. PRIME Lab’s goal was to obtain isotope ratios to a precision of 1% or better given sufficient counting statistics.

The injector and analyzer magnets of PRIME Lab’s tandem accelerator (FN, 8 MV) have large iron cores which are not laminated. Therefore, changes in the magnetic field give rise to large eddy currents within the iron cores. These have a damping effect on the field. For example, prior to this development effort, the analyzer magnet required *ca.* 25–45 sec to stabilize following a 3% step change in current. For a cycle consisting of three magnetic field changes, *ca.* 1.5 min were spent waiting for the magnetic field to settle down. If the radionuclide is counted for 2 min and the stable isotopes are counted for 30 sec each, approximately one-third of the cycle time is wasted.

The acceptable tolerance in the magnetic field is that necessary to achieve the 1% desired precision level. The settling-time is defined as the time taken by the magnetic field to reach and stay within the tolerance band. The injector magnet (before the accelerator) is the most critical since the accelerator has the smallest acceptance of all beam-optical elements. Field changes in this magnet as small as 0.003% (30 ppm) have noticeable effects on measurements; fortunately, this magnet stabilizes much more quickly than the larger analyzing magnet.

A dual controller was designed to meet this objective. The dual controller consists of an optimization algorithm, which is called the settling-time optimizer, and a fuzzy logic controller. The settling-time optimizer reduces the settling-time with every cycle based on the history of previous cycles and the fuzzy logic controller compensates for any disturbances in the system over larger time scales.

**CHLOE: A PROGRAM FOR INTERPRETING *IN-SITU* COSMOGENIC NUCLIDE DATA FOR SURFACE EXPOSURE DATING AND EROSION STUDIES***FRED M. PHILLIPS and MITCHELL A. PLUMMER*

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Cosmogenic nuclides offer many new possibilities for unraveling surficial histories and processes. Although calculations for spallogenic nuclides in simple no-erosion or steady-state erosion cases on flat surfaces are straightforward, many cases of interest involve nuclides produced by low energy neutrons, complex geometries, or combinations of erosion and finite age. We have developed CHLOE (CHLOrine-36 Exposure program), an Excel-based spreadsheet program to facilitate such computations. The program is oriented toward calculations involving  $^{36}\text{Cl}$ , but also incorporates most other commonly-used *in-situ* cosmogenic nuclides. The program uses a database of nuclear parameters to calculate all of the macroscopic low-energy neutron cross-sections used in neutron



flux computations, enabling quantification of the compositionally-dependent neutron fluxes as a function of depth. It also computes background (radioactive-decay induced) neutron fluxes as a function of composition. The program computes scaling factors for the cosmic-ray flux as a function of latitude and altitude. Given data on sample surface angle and topographic shielding it calculates the effects on the surface cosmic-ray flux and attenuation length. As output the program will calculate erosion rates as a function of surface exposure age and will produce multiple-nuclide diagrams. Planned improvements include incorporation of nuclide production by muons and a routine to compute optimal sample size and carrier addition during sample processing.

### **COSMOGENIC EXPOSURE AGE CONSTRAINTS ON TECTONIC HISTORY AND SEMI-ARID LANDSCAPE EVOLUTION**

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Without unequivocal correlation with dated stratigraphic units, determining the timing of last movement along faults is extremely difficult. Cosmogenic nuclides provide the possibility of addressing some of these problems. We have used two *in-situ* cosmogenic radionuclide methods to obtain geochronological evidence in the form of exposure ages to constrain the timing of last movement along three faults bordering Yucca Mountain, Nevada, for the first time. This site is undergoing characterization as a nuclear waste repository. The first method involved the measurement of cosmogenic  $^{14}\text{C}$  in quartz in exposed bedrock fault-line scarps. The 15 measurements, including four from the base of scarps on the Solitario Canyon and Wind Wash faults along the west side of Yucca Mountain, indicate that the scarp has been exposed beyond the saturation value of cosmogenic  $^{14}\text{C}$ , and therefore that no movement had occurred along these scarps in the last 20 kyr. The second method involves the measurement of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in quartz from bedrock knobs, protruding ridges that are crossed by the Ghost Dance fault on the east side of Yucca Mountain. Five preliminary  $^{10}\text{Be}$  data from samples that bound the fault indicate that bedrock on the Antler and Whaleback ridge tops have been exposed for at least 200–680 kyr and 380–1060 kyr. This suggests that the ridge surfaces have been stable since at least the mid-Quaternary and with other data may suggest that there has been considerable time since significant displacement along the fault. These two sites provide a good example of what can be achieved using cosmogenic exposure-age dating to examine tectonic history.

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### **FIRST PERFORMANCE TESTS OF VERA**

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VERA is a new 3-MV pelletron tandem AMS facility in Vienna (see corresponding contribution), which was installed during the last months of 1995 and is currently in the test operation phase. The goal of this phase is to establish the conditions for performing AMS measurements of  $^{10}\text{Be}$ ,  $^{14}\text{C}$  and  $^{26}\text{Al}$ .

The present report will discuss the performance characteristics of the facility established so far, and present first measurements.

## THE INFLUENCE OF ARCHAEOLOGICAL CONTEXT ON THE STUDY OF MARINE RESERVOIR AGE VARIATION

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Radiocarbon age calibration for marine samples requires the application of a reservoir correction. The magnitude of this correction depends on the regional balance between upwelling and ocean atmospheric CO<sub>2</sub> exchange and is important for both paleoenvironmental and archaeological applications. To account for regional differences in reservoir age, Stuiver *et al.* (1986) define a term called delta-R. The  $\Delta R$  suggested for southern California,  $225 \pm 35$ , is averaged from seven samples located between Bolinas Bay and San Diego (Stuiver *et al.* 1986). Because offshore depth, currents, and upwelling, among other factors, contribute to local variation in the reservoir effect, accurate <sup>14</sup>C age calibration requires a site-specific correction factor.

In this study, comparison of three archaeological deposits from southern California illustrate the complexities surrounding the estimation of marine reservoir age corrections and the calculation of a local  $\Delta R$ . Fourteen shell-charcoal pairs were recovered from archaeological sites along the Newport Coast. AMS <sup>14</sup>C analysis was necessary to maximize the sampling context given the small specimen size. Wood charcoal remains were removed from inside each shell. The AMS results indicated that the  $\Delta R$  obtained for each set appeared to be dependent on the formation of the archaeological deposit. The study implied that samples recovered from an open site lacking vertical strata were more accurate. In contrast, samples recovered from dense rock-shelters with stacked cooking areas contained shell-charcoal remains that were not contemporaneous and thus less appropriate for calculating  $\Delta R$ .

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## A NEUTRAL BEAM AMS SYSTEM FOR ULTRASENSITIVE DETECTION OF ATOMS THAT DO NOT FORM NEGATIVE IONS

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In a companion paper, Litherland and Kilius discuss the usefulness of neutral beam injection for the AMS detection of atoms, such as krypton, that form negative ions with difficulty or not at all. A feature of neutral injection is that within the drift region between ground and the high voltage terminal, where the polarity of such particles is converted from neutral to positive, the trajectories are unaffected by the electric field. Consequently, the direction of the electric field can be strongly angled to the path of the neutral beam, causing any unwanted secondary particles to be removed within a few centimeters of their formation point. Thus, the normal voltage gradient limitation of long tubes,

known as the total voltage effect, can be avoided allowing the reliable use of very high electric fields. With proper surface coatings, electric fields above 5MV/meter appear to be practical, allowing the neutral beam drift distance to be short and the optical magnification between the virtual source of the neutral particles and a beam cross-over at the center of the terminal charge changing canal or jet, to be close to unity. A relevant factor is that in such devices high vacuum, rather than high pressure sulfur hexafluoride gas, may become very desirable for high voltage insulation. Apart from the very high voltage gradients that become usable, vacuum insulation allows the elimination of the triple point between glass, vacuum and electrode together with radial pumping of gas molecules that exit from the terminal stripper or jet rather than axial along the tubes. A description will be presented of a proposed vacuum insulated neutral beam AMS instrument for  $^{81}\text{Kr}$  detection.

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## AMS DETECTION OF CHLORINE-36 USING SMALL TANDEMS

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The most significant interference that is present during AMS detection of  $^{36}\text{Cl}$  atoms is the stable isobar  $^{36}\text{S}$ . Sulfur readily forms negative ions and it has been found that the ubiquitous sulfur contamination that is present within almost all samples produces a background  $^{36}\text{S}$  events that cannot be completely eliminated by any practical combination of electric and magnetic filters. It has been found that to achieve useful  $^{36}\text{Cl}$ - $^{36}\text{S}$  isobaric separation, it is necessary to not only take great care during sample preparation to eliminate sulfur contamination but also to accelerate the ions to energies of at least 30MeV before they are directed into an ionization detector for element identification. Achieving such final energies requires the use of large nuclear physics-type tandems capable of operating at terminal voltages above 5MV. The present paper describes a new detection system which avoids this requirement. The principles are as follows: A small tandem accelerates wanted ions in the 4+ charge state to an energy of 15MeV. At this energy, the ions are post-acceleration analyzed and stripped to select only mass-36 particles in the 7+ charge state. Following this selection, the ions are directed into a conventional ionization detector located within a high voltage terminal that is maintained at an electric potential of -3MV. Thus, 21MeV is added to the energy of the ions after the previous analysis for a final energy of 36MeV. Such a detection arrangement uses well-established detection and high voltage generation technology. This new detector can be designed to be compact and operate reliably with little maintenance.

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## HYBRID ION SOURCES: RADIOCARBON MEASUREMENTS FROM MICROGRAM TO MILLIGRAM

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Gas ion sources provide a very efficient way of dealing with very small samples and, by injecting samples in a stream of helium carrier gas, measurements can be obtained from as little as 1  $\mu\text{g}$  carbon. This is particularly useful for measurements on high activity samples and for the study of sample contaminants in relation to high precision AMS measurements. Another benefit of gas based measurements is that they can be made in real time and we discuss the range of possible applications

such as GC-AMS and LC-AMS which could make AMS more widely applicable in the life sciences. Graphite ion sources provide a more efficient use of accelerator time for the measurement of large samples because of the higher carbon currents attainable. The precisions attainable in this way are so good that re-evaluation of possible sources of systematic error (such as low level contamination and sample size effects) is essential. Hybrid ion sources capable of operating on either gas or graphite allow the advantages of both techniques to be exploited. With minor modifications to the gas ion source at Oxford we are now able to run on both sample types. Currents from gas are typically 10–12  $\mu\text{A}$  and those from graphite 40–50  $\mu\text{A}$ . Maximum currents from graphite exceed 300  $\mu\text{A}$ . We discuss the implications of this for the future development of radiocarbon AMS facilities designed for a wide variety of measurements and research.

**APPLICATION OF ANTHROPOGENIC  $^{129}\text{I}$  AS A TRACER OF NUCLEAR EMISSIONS: A STUDY AROUND POTENTIAL POINT SOURCES AT A NUCLEAR FUEL REPROCESSING PLANT AND TWO NUCLEAR POWER PLANTS IN UPSTATE NEW YORK**

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We have investigated the presence of anthropogenic  $^{129}\text{I}$  in upstate New York where several potential point sources of this long-lived radioisotope exist. For the study samples were collected of surface waters, soils and biological materials around the nuclear fuel reprocessing facility at West Valley near Buffalo (operated between 1966–1972), and near two active nuclear power plants at Ginna near Rochester and at Nine Mile Point near Oswego, both along the Lake Ontario shoreline, as well as at several other sites in and outside upstate New York.

A striking signal of  $^{129}\text{I}$  levels of  $10^{11}$  atoms/L of discharge was detected just outside the West Valley site boundary in Buttermilk Creek which drains the site. This signal can be tracked into Lake Erie via Cattaraugus Creek which receives Buttermilk Creek before flowing into Lake Erie at Silver Creek. Levels in other creeks in the region of the reprocessing plant were on the order of  $10^9$ – $10^{10}$  atoms/L, and showed marked directional trends that could be correlated with predominant wind directions at the time of minor releases at the site. Levels of  $^{129}\text{I}$  in surface waters decreased with distance from West Valley and were constant at  $10^8$  atoms/L for the entire upstate New York area outside a 50-mile radius of the site, including around the two nuclear power plants in the region.  $^{129}\text{I}$  levels in surface waters outside upstate New York in areas such as the Adirondacks region of eastern New York, Maine, Vermont, Canada, *etc.* are on the order of  $10^7$  atoms/L. Although these concentrations are an order of magnitude lower than levels in upstate New York, they are still elevated by over three orders of magnitude above pre-nuclear values. Waters and plants around an active nuclear power plant in Ohio also show  $^{129}\text{I}$  levels within this same order of magnitude as other non-point source locations outside upstate New York.

Soil, grass and aquatic plant samples collected from these various locations and were prepared for AMS measurement by alkali leach/fusion extraction and subsequent extraction into carbon tetrachloride. This data also show similar spatial trends, with highest levels around West Valley. Some data for aquatic plant/water and grass/soil bioconcentration factors will also be presented. As part of the same study, we made some pilot measurements of  $^{129}\text{I}$  levels in tree-rings obtained from trees at West Valley and at Rochester.

Data obtained so far suggest that ring-porous woods such as elm, maple and locust are better indicators of  $^{129}\text{I}$  preservation in annual tree rings than are diffuse-porous woods such as Cherry. The limiting of the water-conducting tissue, xylem, to the outermost annual ring in ring-porous woods appears to minimize transport of iodine across tree rings via the hydrologic system, in contrast to diffuse-porous woods in which the xylem is distributed through all the growth layers of the tree.

$^{129}\text{I}$  levels in all the above reservoirs, *i.e.*, waters, aquatic vegetation, grass, soil, and trees in the West Valley region from this study and from other workers were all compared with total  $^{129}\text{I}$  estimated to have been released from the site during its operation. Despite a striking signal of  $^{129}\text{I}$  outside the site boundary over two decades after the site was closed, the total  $^{129}\text{I}$  around the site is in fact only a very small fraction of this radionuclide released at the site. This indicates that most of the  $^{129}\text{I}$  has migrated off-site by this time, leading to a signal that can be tracked to the Great Lakes and also causing a stronger presence of this isotope in upstate New York than elsewhere.

### CHEMICAL PREPARATION OF ENVIRONMENTAL WATER AND BIOLOGICAL SAMPLES FOR THE DETECTION OF ANTHROPOGENIC $^{129}\text{I}$ BY AMS

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We have carried out an analysis of anthropogenic  $^{129}\text{I}$  levels in upstate New York where an inactive nuclear fuel reprocessing facility and two active nuclear power plants are housed. Recent  $^{129}\text{I}$  produced since the onset of the nuclear era has found its way into all reservoirs in exchange with the atmosphere, and has led to elevated  $^{129}\text{I}/\text{I}$  ratios there. In order to evaluate levels of this isotope around potential point sources in the study area, we prepared samples of surface waters, aquatic vegetation, soils, grasses and wood collected around these specific sources and other locations for background comparison for AMS analysis.

Rainwater and surface waters collected from creeks and lakes have extremely low concentrations of iodine averaging  $0.2 \mu\text{g}/\text{L}$  for >70 samples measured. This necessitated the vacuum distillation of one to two liters of water for each sample down to a few hundred milliliters, to increase iodine concentration in the samples to levels where detection by ion chromatography was possible. The original iodine concentration in each sample was then calculated from the pre- and post-distillation sample volumes, and the concentration after distillation. Carrier iodine of known  $^{129}\text{I}/\text{I}$  ratio and iodine concentration was added to the sample after vacuum distillation in order to increase the bulk of the sample and to eliminate contamination of the ion source by hot samples, *i.e.*, those high in  $^{129}\text{I}$ , during AMS measurement. The sample and carrier was extracted into carbon tetrachloride after conversion to periodate to achieve isotopic equilibrium and then back-extracted into the aqueous phase following standard procedures (see Fehn *et al.* 1992).

Biological samples such as vegetation and soils have much higher concentrations of iodine, typically  $1\text{--}5 \text{ mg kg}^{-1}$ , yet pose a different challenge in the form of extracting iodine successfully from organic materials. After experimentation with methods such as oxygen combustion in a Parr oxygen bomb, an alkali leach/fusion extraction was chosen as the best of available alternatives for extracting iodine from biological materials. This process is a modified version of one originally utilized by K. Nishiizumi (1982) for meteorite samples with successive adaptations by J. Fabryka-Martin (personal communication) and others. Sample size required usually varies from 1 to 10 mg, depending on concentration of iodine in sample type and the amount of carrier iodine which could be added which in turn depends on the expected  $^{129}\text{I}/\text{I}$  ratio in the sample. Saturated sodium hydroxide was

added to dried samples in previously unused metal crucibles. This mixture was thoroughly mixed and dried for 24 hr under a heat lamp. Sodium peroxide was added to the samples to break down the organic molecules. The sample crucibles were subsequently heated in a furnace for two hours at temperatures of 600°C. A 50/50 mixture of  $^{2}\text{N}$  sulfuric acid and sodium bisulfite was added to the fluxed sample, to reduce iodine present to iodide. Carrier iodine was added at this stage and extraction was completed following the same procedure as for the water samples. In the case of a few test samples and three soil samples obtained from NIST (non-certified), carrier iodine was added prior to fluxing the samples in order to monitor recovery rates during fluxing, which typically varied between 40–70%.

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## FIRST $^{14}\text{C}$ OBSERVATIONS IN WATERS OF THE GREAT AUSTRALIAN BIGHT

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The Great Australian Bight (GAB) to the south and the Indonesian Passage to the north of Australia are both important passages for the flow of surface, intermediate and deep water between the Indian Ocean and Pacific Ocean. Both field observations and ocean models have clearly identified the role of the Indonesian Passage for the global thermohaline circulation. In contrast, less well understood is the flow pattern south of Australia; both in direction as well as in absolute quantities.

$^{14}\text{C}$  is a tracer of this flow and for the first time, water samples for subsequent  $^{14}\text{C}$  analysis were collected at several locations in the Bight during the RV *Franklin* cruise of July 1994. The first samples from two profiles in the eastern domain of the GAB were analyzed recently and we report and discuss the observed  $^{14}\text{C}$  distribution in relation to the general hydrography of the region.

For all sample depths, a close agreement of the data with previous measurements obtained during the international Geochemical Ocean Sections Study (GEOSECS) from the southeast Indian Ocean was found. We obtained  $\Delta^{14}\text{C}$  values for surface, intermediate and deep water with approximately 90‰, –100‰ and –150‰, respectively. The minimum concentration found at 200-m depth coincides with a subsurface minimum observed during the GEOSECS expedition. The value is most likely representative of newly formed Sub-Antarctic Mode Water.

Our data represent the first successful Australian AMS Spectrometry measurements for ocean water samples with a precision of <0.5%. This high precision is required to identify the weak horizontal gradients of the natural radiocarbon distribution within the deep ocean.

The GAB  $^{14}\text{C}$  data link the Indian Ocean and Pacific Ocean data bases established within the framework of the World Ocean Circulation Experiment (WOCE). Our measurement program of  $^{14}\text{C}$  in

Australian ocean waters is now established, allowing commencement of the analysis of WOCE samples at ANTARES.

### **CHOLESTEROL TURNOVER AND METABOLISM STUDIES IN HUMAN SUBJECTS: THE ROLE OF OXYSTEROLS**

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Cholesterol absorption, turnover and metabolism in the human body is a subject of great interest to the medical profession. Oxysterols, the products of cholesterol oxidation, can have detrimental effects (many investigators believe that they are a cause of atherosclerosis), but have potentially desirable effects, in that they seem to mediate the actions of cholesterol. Cholesterol circulates in the body in relatively large quantities, but oxysterols do not. Concentrations of labeled oxysterols become unmeasurable by conventional techniques in a few minutes.

AMS provides the capability of conducting studies of the action on cholesterol of oxysterols. Physiologically trivial amounts of  $^{14}\text{C}$ -labeled cholesterol and 7- $\alpha$ -hydroxycholesterol can be injected into human subjects, and their concentrations in plasma followed for several days. The sterols are extracted from plasma, and purified by chromatography. The two fractions are then analyzed by AMS at PRIME Lab. Techniques for handling extremely small samples of oxysterols are emphasized. Preliminary results of studies will be presented.

### **THE LLNL AMS FACILITY**

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The AMS facility at Lawrence Livermore National Laboratory routinely measures the isotopes  $^3\text{H}$ ,  $^7\text{Be}$ ,  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ,  $^{63}\text{Ni}$ , and  $^{129}\text{I}$ . During the past year, over 15,000 research samples have been measured. Of these samples, approximately 30% were biomedical  $^{14}\text{C}$  tracer samples, 40% were  $^{14}\text{C}$  samples for archaeology and the geosciences, with the remaining 30% for the other isotopes. During the past two years, a significant amount of work at LLNL has gone into the development of the Projectile X-ray AMS (PXAMS) technique. PXAMS uses induced characteristic X-rays for discrimination of competing atomic isobars. PXAMS has been most fully developed for  $^{63}\text{Ni}$  and shows promise for the measurement of several other long lived isotopes. During the past year LLNL has also conducted an  $^{129}\text{I}$  interlaboratory comparison exercise.

Recent hardware changes to our facility include the installation and testing of a new thermal emission ion source, a new multi-anode gas ionization detector for general AMS use, a new cryo-vacuum system for the AMS ion source, and the re-alignment of the vacuum tank of the first of the two magnets that make up the high energy spectrometer. In addition, we have begun design studies and carried out tests for a new high-resolution injector and a new beamline for heavy element AMS.

This work was performed under the auspices of the U.S. Department of Energy at the Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

### **<sup>129</sup>I INTERLABORATORY COMPARISON**

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An interlaboratory comparison experiment for <sup>129</sup>I has been organized and conducted. A total of six laboratories participated in the exercise to either a full or limited extent. In the comparison, a suite of 11 samples were used. This suite of standards contained both synthetic "standard type" material (e.g., AgI) and environmental materials. The isotopic <sup>129</sup>I/<sup>127</sup>I ratio of the samples varied from 10–8 to 10–14. Results of the comparison are presented.

This work performed under the auspices of the U.S. Department of Energy at the Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

### **DEVELOPING A <sup>14</sup>C-TARGET PREPARATION SYSTEM FOR THE LEIBNIZ-LABOR AMS-FACILITY**

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At the Leibniz-Labor für Altersbestimmung und Isotopenforschung of the Christian-Albrechts-University a target preparation system for AMS <sup>14</sup>C measurements is under construction. The planning is to produce CO<sub>2</sub> from organic material with a closed-quartz-tube combustion and from inorganic material by hydrolysis with phosphoric acid. The CO<sub>2</sub> will be converted to graphite with the standard Fe/H<sub>2</sub> catalyzed reduction and then pressed into an aluminum holder. For the reduction we will install manifolds with 10–12 reduction units. For the target preparation systems several factors are considered, namely: 1) the effect of the amount of carbon and of the Fe/C mixing ratio on current intensity, sample endurance, isotope ratio and background; 2) the effect of pretreatment and particle size of the Fe catalyst on reduction rate and background; and 3) and background contamination incurred during sample preparation (presently 40,000–45,000 yr apparent <sup>14</sup>C age for a "dead" foraminifer carbonate). We will report and discuss results obtained thus far.

### **DEEP WATER FORMATION AND EXCHANGE RATES IN THE ARCTIC OCEAN: IMPLICATIONS FROM THE DISTRIBUTION OF <sup>14</sup>C**

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We present and discuss a <sup>14</sup>C data set covering all major basins of the Arctic Ocean (Nansen, Amundsen, Makarov and Canada basins). The data were collected during two cruises of RV *Polarstern* to the Nansen, Amundsen and Makarov basins of the Arctic Ocean (ARK IV/3, 1987, and Arctic 91 expedition), and a cruise of the Canadian icebreaker CCGS *Louis St. Laurent* to the Canadian Basin (Arctic Ocean Section 94). The data set consists of ca. 250 large-volume samples measured radiometrically at the University of Heidelberg and ca. 150 small-volume samples measured at the Woods Hole AMS facility. δ<sup>14</sup>C values of bottom waters reach from ca. –75‰ in the Amundsen Basin to ca. –105‰ in the Makarov and Canada basins. δ<sup>14</sup>C values of the bottom waters



in the Nansen Basin fall between these two values ( $-85\text{‰}$ ). The  $\delta^{14}\text{C}$  values of the bottom waters in the Makarov and Canada basins are remarkably constant. There is no detectable gradient below *ca.* 2000 m across the entire Canadian Basin. The bottom waters of the Eurasian Basin (Nansen and Amundsen basins) contain traces of bomb tritium. The bomb tritium is used to correct the measured values for bomb  $^{14}\text{C}$ . Mean “ages” of bottom waters derived from the corrected  $\delta^{14}\text{C}$  values range from *ca.* 250 to 300 yr in the Eurasian Basin to *ca.* 350 to 400 yr in the Makarov Basin. Simple time-dependent tracer balances (box models) are used to convert these “ages” into mean deep-water formation and exchange rates.

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### DETERMINATION OF PROTON-INDUCED PRODUCTION CROSS SECTIONS AND PRODUCTION RATES OF $^{129}\text{I}$ FROM Te IN THE MEDIUM ENERGY RANGE

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The radioisotope  $^{129}\text{I}$  ( $t_{1/2} = 1.57 \times 10^7$  yr) can be employed to examine the constancy of the galactic cosmic ray (GCR) flux on longer time scales than by determination of  $^{53}\text{Mn}$  ( $t_{1/2} = 3.7 \times 10^6$  yr). In extraterrestrial material, Te is one of the main target elements for the production of  $^{129}\text{I}$  (mainly by secondary neutrons).

In the course of a systematic investigation of proton-induced reactions with different target elements, production cross sections of  $^{129}\text{I}$  from Te were determined after irradiation of samples of  $\text{Cu}_2\text{Te}$  (500 mg Te) with doses varying from  $1.9 \times 10^{14}$  to  $1.2 \times 10^{15}$  protons  $\times \text{cm}^{-2}$  at six different energies of the incident beam between 330 and 1600 MeV at Laboratoire Nationale Saturne (LNS), Saclay. Cross sections as determined in these experiments are the basic input data of model calculations of production rates and provide the means to validate these.

As part of experiments to simulate the production of cosmogenic nuclides by GCR protons, samples of Te were placed at different depths in artificial meteoroids made of gabbro (“stony meteoroid”, dose:  $1.3 \times 10^{14}$  protons  $\text{cm}^{-2}$ ) (Michel *et al.* 1995) and iron (“iron meteoroid”, dose:  $2.17 \times 10^{14}$  protons  $\text{cm}^{-2}$ ) (Michel *et al.* 1993), respectively, were irradiated isotropically with 1600 MeV protons at LNS, Saclay, to determine the production rates of  $^{129}\text{I}$  from Te. Production rates derived from such simulation experiments can be used to validate the production rates of nuclides produced by GCR particles because of the known irradiation conditions in the simulation experiments.

Iodine was separated from the irradiated samples by digestion with NaOH after addition of iodide and iodate carriers of low isotopic  $^{129}\text{I}/^{127}\text{I}$  ratio. In the case of  $\text{Cu}_2\text{Te}$ , sodium thiosulfate was added to avoid formation of iodine and precipitation of  $\text{CuI}$ . After reduction to iodide using  $\text{HSO}_3^-$ , Te was removed by extraction of iodine into chloroform after oxidation by  $\text{NaNO}_2$ . Reduction to iodide and re-extraction into water was followed by precipitation of  $\text{AgI}$ . The  $^{129}\text{I}/^{127}\text{I}$  ratios (on the order of  $10^{-10}$ ) were measured at the ETH/PSI tandem accelerator using 4.7 MV on the terminal and charge state  $5^+$  from the gas stripper. A time-of-flight spectrometer was used to suppress background from traces of Te in the samples.

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## KINEMATIC EFFECTS IN THE SPUTTERING OF CARBON ISOTOPES

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Isotopic data accumulated by AMS on sputtered carbon from processed and unprocessed graphite has revealed effects due to the presence of the iron catalyst. These effects have been noted before, in the literature, but never quantified. In opposition to the usual preferential sputtering of the lighter isotope at small angles, we see a small enhancement of the heavier isotope, when the iron catalyst is present. This amounts to *ca.* 5% higher  $\delta^{13}\text{C}$  values than those for pure graphite. We have attempted to model this effect using molecular dynamics calculations. We used a simple representation for the graphite and iron lattices, and assumed that the iron is uniformly coated with graphite. Results indicate that the fractionation differs with lattice orientation, and that the carbon ions show extended emission from the iron-containing lattice. The implications for AMS analysis will be discussed.

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## THE PRIME LAB EXTERNAL RESEARCH PROGRAM

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PRIME Lab is a national facility supported primarily by the Geosciences Directorate of the National Science Foundation for measurement of the long-lived cosmogenic nuclides  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ , and  $^{129}\text{I}$ . Since its inception in 1991 (when we started our external research) we have measured *ca.* 4500 samples and have chemically prepared *ca.* 2000 samples. Currently there are a total of *ca.* 110 external users conducting research using AMS data obtained at PRIME Lab. There are *ca.* 160 ongoing external research projects. *Ca.* 60% of the samples measured at PRIME Lab belong to the external research program and *ca.* 40% of these are prepared in the PRIME Lab chemistry laboratory. We have a special seed analyses program to attract new users and to allow them to obtain preliminary data for proposals.

Currently the external research program is very multidisciplinary. Areas of research for the external program are atmospheric science, ocean science, hydrogeology, planetary science, glacial chronology and geomorphology, soil science, archaeology, anthropology, environmental science and biology. *Ca.* 50% of the research projects are centered around applications of *in-situ*-produced cosmogenic nuclides ( $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$ ) for geomorphology and climate change. Applications of *in-*

*situ*-produced nuclides include studying glacial chronology, erosion and paleolake shorelines; determining tectonic uplift rates and seismic hazards; dating of landslides, volcanic lava flows, rock art and paleoearthquake events.

Types of other research applications include: radionuclide fallout and mapping over the United States; soil dynamics and chronology; cosmic ray intensity variations; digenesis of ferromanganese crusts; distribution of  $^{129}\text{I}$  and carbon cycling in the oceans; groundwater tracing; assessment of low level and high level radioactive waste repositories; release of radioactivity from fuel reprocessing facilities and reactors; toxicity and uptake of Al by plants and animals; determination of neutron energy spectra from atomic bombs at Hiroshima and Nagasaki; exposure histories and terrestrial ages of the meteorites and tektites.

### APPLICATION OF $^{36}\text{Cl}$ SURFACE EXPOSURE AGE DATING TO CENTRAL ANDEAN VOLCANOLOGY AND GLACIOLOGY

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The Central Andes (14–27°S) is characterized by high altitude (~4000 m), arid climate (precipitation <20 cm a<sup>-1</sup>), extremely low erosion rates (~1 mm ka<sup>-1</sup>) and superb preservation of the geology. Here we describe two ongoing research projects that utilize *in-situ*-produced cosmogenic  $^{36}\text{Cl}$  as a late Pleistocene chronometer to better understand the magmatic evolution and quaternary paleoclimate of this region.

The Central Volcanic Zone of the Andes is probably one of the best preserved young volcanic regions in the world. Studying the magmatic processes of this region will help us to better understand magmatism and geodynamics at such margins globally. Our objectives are to: 1) constrain production rates of  $^{36}\text{Cl}$  and calibrate  $^{36}\text{Cl}$  exposure ages using high precision  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages on the same lava flows; 2) estimate rates of growth and evolution of volcanoes; 3) estimate regional magma generation rates; and 4) compare radiometric ages with the remotely sensed geomorphological data. We have completed preliminary sampling of Volcan Tata Sabaya in southwest Bolivia as it is <1 Ma old and its volcanic and petrologic evolution is well known. A total of 30 samples have been collected from lava flows and debris avalanche blocks for age dating using  $^{36}\text{Cl}$  and  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  methods. Analytical work on recently collected samples is in progress. During the coming summer we intend to complete sampling at Tata Sabaya and sample the Volcan Parinacota, another major volcano in this region.

Quantitative estimates of past climates are required to better address global climatic changes and evaluate the predictive capabilities of general circulation models. We have initiated a study of the glacial landforms in the mountains of the Western Cordillera and Altiplano of Bolivia to decipher the late Quaternary glacial record in the tropical regions. Three well preserved moraine sets have been identified on the basis of superposition and terminus elevation at Cerro Tunupa. Surface exposure ages have been determined using  $^{36}\text{Cl}$ . Three main periods of glacial advance are suggested. An advance during isotope stage 3 is suggested by the 43 ka age obtained from the oldest moraine set (Group III). The dates spanning from 32–15 ka for moraine set II corresponds to advance during isotope stage 2 Late Glacial Maximum. Boulders from the lateral moraine on the youngest and most extensive glacial valley yields ages of 14–12 ka corresponding to a late glacial advance. These data indicate a close correspondence between the neoglacial and paleolake history of the Altiplano and will provide constraints to central Andean paleoclimate reconstruction.

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## A PRELIMINARY REPORT ON THE CHARACTERISTICS OF A CO<sub>2</sub> GAS ION SOURCE MGF-SNICS/SIMULTANEOUS INJECTOR AT NIES-TERRA

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The accelerator mass spectrometer at the National Institute for Environmental Studies (NIES-TERRA; Tandem accelerator for Environmental Research and Radiocarbon Analysis) has two ion source-injector combinations, *i.e.*, the combination of multi-cathode Cs sputter source for solid samples (MC-SNICS) with a fast bouncing sequential injector, and that of multiple gas feed negative ion source (MGF-SNICS) with a simultaneous injector dedicated to the carbon isotope analysis of gaseous samples. The MGF-SNICS (Ferry, in press) was developed based on the SNICS-II source with influences from the existing gas sources (Bronk and Hedges 1987; Middleton, Klein and Fink 1989). The target is composed of a piece of high purity (99.999%) Ti mounted in a small aluminium shell. CO<sub>2</sub> sample is fed from a 1.5 ml stainless steel cylinder through a 75-cm-long stainless steel tube. The gas flow rate is controlled by a metering valve attached on a side of each cylinder. The source has twelve independent sets of the above target-tube-valve/cylinder complex in a target magazine assembly to minimize the chance of cross contamination between the samples.

The negative carbon ions extracted from the MGF-SNICS are separated and then combined by the two magnetic analyzers and three electrostatic lenses for the simultaneous detection of <sup>14</sup>C, <sup>13</sup>C and <sup>12</sup>C isotopes after acceleration. The basic design of the simultaneous injector is influenced by the system developed by Southon *et al.* (1990). Combination of a gas ion source with a simultaneous injector is expected to effectively suppress space charge effects (Bronk Ramsey and Hedges 1994) caused by the presence of large amount of O<sup>-</sup>.

Preliminary data on the performance of MGF-SNICS will be reported.

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### MICROBEAM AMS: PROSPECTS OF NEW GEOLOGICAL APPLICATIONS

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AMS applications in geology have hitherto concentrated on the use of cosmogenic isotopes and rare *in-situ* produced isotopes for geomorphological and geophysical studies. Special features of AMS lend themselves to more general applications to other isotopes and geochronological systems. Advantages of *in-situ* microanalytical techniques over bulk techniques have been demonstrated not only in terms of expedience but also in allowing analysis of microscopic geological samples (*e.g.*,

inclusions) or micro-features of minerals (*e.g.*, zoning) not amenable to bulk methods. *In-situ* measurements in geochronology has been carried out with ion-microprobes, but isobaric and molecular mass interferences restrict the scope to special systems where the problem is minimal. AMS can be used to alleviate this mass interference problem, and opens up the prospect of a less restrictive *in-situ* microanalysis for geochronology. At CSIRO, a microbeam AMS system designed to achieve this capability is under construction. With this system, several interesting applications such as the Re-Os system became accessible more conveniently. The U-Pb system becomes accessible for hydrous minerals, and Rb-Sr systems for Rb rich samples. In addition, AMS allows determination of trace elements at lower level than those accessible with the proton microprobe. The paper will discuss these prospects and describe the AUSTRALIS system (AMS for Ultra Sensitive TRAc eLement and Isotopic Studies) being developed at CSIRO.

### THE AUSTRALIS MICROBEAM ION SOURCE

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The AUSTRALIS (AMS for Ultra Sensitive TRAc eLement and Isotopic Studies) system being developed at the HIAF laboratory is a microbeam AMS system designed for *in-situ* microanalysis of geological samples for ultra-trace (sub-ppb) and isotopic data. In the first stage of the development now completed, the microbeam source was implemented by modifying a HICONEX source. The modification results in a versatile source that can be used both for high intensity macrobeam applications, as well as microbeam. The sample chamber features a sample viewing system at normal takeoff angle, in the reflected geometry, enabling live observation of the sputtering process and visual tuning of the microbeam. A microbeam of Cs<sup>+</sup> as small as 30 microns in diameter has been obtained in the tests. The secondary ion extraction system features a "screen" electrode that is used to correct the microbeam trajectory affected by the secondary ion extraction field, in order to return it to the geometric center of the sample. The paper will describe the source, sample chamber and results of the test.

### A FAST ISOTOPE SWITCHING SYSTEM FOR HIGH ENERGY IONS

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To achieve the high precision required for isotopic ratios in geochronological applications of AMS, a fast isotope switching system has been implemented for the AUSTRALIS (AMS for Ultra Sensitive TRAc eLement and Isotopic Studies) system being developed at the HIAF laboratory. Slow drifts in the accelerator stability as well as in beam transport components dictates that the sequence of isotopes of interest should be counted for as short an interval as possible. For a narrow range of transmission typical for most analyzing magnets, fast switching (<1 ms) of the field is not possible if steady state is also desired. We have thus devised an electrostatic means to modulate the beam trajectory in a static magnetic field. The system is based on a pair of deflector plates sets, deflecting in the orbit plane, at the entrance and exit of the high energy analyzing magnet. Originally designed as an energy modulator, the principle readily adapts as an isotope switcher. The paper will discuss the design of the system and its performance.

**QUANTIFICATION OF TRACE ELEMENT ANALYSIS BY AMS USING PIXE**

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There is increasing recognition, matched by increased development effort, of the ultra-trace detection capability of AMS, especially in materials science and geological applications. The destruction of molecular ions by Coulomb explosion after the charge-changing collisions in the accelerator terminal simplifies the detection of many elements, even at sub-ppb level. Quantification of the results, however, still presents a challenging problem, arising mainly from matrix effects of the sputtering process. For a simple matrix, *e.g.*, silicon, standards can be prepared to solve the problem. In a complex matrix, *e.g.*, typical geological samples, the challenge can be quite formidable. Matrix effects include compositional and conductivity effects. The concentration of the trace elements of interest in the standards cannot be made too high to avoid non-linearity problems over too large a dynamic range. PIXE is an ideal technique for characterizing standards with trace elements in the ppm range, allowing a good range of overlap of the sensitivity ranges of AMS and PIXE. Prospects and an example in PGE calibration in meteorites will be discussed.

**MEASUREMENT OF PROTON PRODUCTION CROSS SECTIONS OF  $^{10}\text{Be}$  AND  $^{26}\text{Al}$  FROM ELEMENTS FOUND IN LUNAR ROCKS**J. M. SISTERTSON,<sup>1</sup> K. KIM,<sup>2</sup> P. A. J. ENGLERT,<sup>2</sup> M. CAFFEE,<sup>3</sup> C. CASTANEDA,<sup>4</sup> J. VINCENT<sup>5</sup> and R. C. REEDY<sup>6</sup>

Long-lived radionuclides are produced in extraterrestrial materials from cosmic ray interactions. From this cosmogenic archive the exposure history of the object can be characterized. In many instances, the goal of cosmogenic radionuclide measurements is to determine the duration of exposure to cosmic rays of the object as a meter-sized body. In some instances, the use of several cosmogenic radionuclides can be used to constrain complex exposure histories. Alternatively, the precise measurement of several radionuclides in extraterrestrial materials can be used to estimate the solar proton flux over a time frame comparable to the half-lives of the radionuclides (Reedy and Marti 1991). In this environment, 98% of solar cosmic rays and ~87% of galactic cosmic rays are protons, so the cosmogenic archive can be analyzed if, and only if, the proton production cross sections are well known. Although the development of Accelerator Mass Spectrometry (AMS) has made it possible to perform radionuclide measurements readily, many of the relevant cross sections have never been measured or only a single measurement is available for a particular target material (see for example Sisterson *et al.* 1994; Bodemann *et al.* 1993).

$^{10}\text{Be}$  and  $^{26}\text{Al}$  production from carbon, silicon, aluminum, magnesium and quartz targets have been measured over a proton energy range of ~30–500 MeV using three accelerator facilities for the target irradiations. To minimize both proton scattering out of the stack and secondary neutron production within the stack, thin targets with “catcher” foils are used. At most, ~2 MeV is lost in any target and ~10 MeV in the entire stack. A transmission ion chamber or secondary electron emission monitor calibrated with a Faraday cup for each irradiation energy, gives a direct measure of the number of protons through the target stack. Monitor foils are used as an additional dosimetry check. Irradiation times were optimized to produce  $10^7$ – $10^8$   $^{14}\text{C}$  atoms in each target (Sisterson *et al.* 1994). At most energies, these irradiation times produced sufficient  $^{10}\text{Be}$  and  $^{26}\text{Al}$  atoms in the target for analysis to be possible; the chemistry for  $^{10}\text{Be}$  and  $^{26}\text{Al}$  was performed at San Jose State University and the AMS measurement made at Lawrence Livermore National Laboratory AMS facility (Southon *et al.* 1990).

We report new measurements for the cross sections  $O(p,x)^{10}\text{Be}$ ,  $C(p,x)^{10}\text{Be}$ ,  $Si(p,x)^{10}\text{Be}$ ,  $Mg(p,x)^{10}\text{Be}$ ,  $Al(p,x)^{10}\text{Be}$ ,  $Si(p,x)^{26}\text{Al}$  and  $Mg(p,x)^{26}\text{Al}$ . These values and values for the  $O(p,x)^{10}\text{Be}$  cross section reported before (Sisterson *et al.* 1992), will be compared to those in the literature and implications for solar proton fluxes over the time period characterized by the radionuclides  $^{10}\text{Be}$  and  $^{26}\text{Al}$  considered.

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## THE ACTINIDES MEASUREMENT PROGRAM AT ANTARES: INITIAL TESTING OF THE NEW HIGH ENERGY, SPHERICAL ELECTROSTATIC ENERGY ANALYZER

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A major goal at ANTARES is to develop a program for measuring actinides with AMS. We plan to develop techniques for detecting rare radionuclides such as  $^{236}\text{U}$ ,  $^{229}\text{Th}$ ,  $^{230}\text{Th}$ , and  $^{244}\text{Pu}$  at natural levels, utilizing a purpose-built beam line. In addition, we plan to investigate the advantages of AMS for measuring isotopic ratios for major isotopes at low concentrations directly in the original matrix.

The “actinides beamline” will be constructed from all new components, using electrostatic lenses for mass independent focusing. Following the ideas of Purser (in press), an electrostatic quadrupole doublet immediately following the accelerator will focus the beam at a gas post stripper, where the second stripping to high charge states will allow us to 1) bend high masses at higher energies with an affordable magnet, and 2) reject molecular fragments having similar  $m/q$ . A consequence of this approach is that the electrostatic quadrupole will have to be capable of focusing charge state  $1+$  ions at energies up to 16 MeV. Following the post stripper, another electrostatic doublet will transport the beam through the existing analyzing magnet to the object of a  $90^\circ$  spherical electrostatic analyzer (ESA). Following the ESA will be a new analyzing magnet with resolving power similar to the ESA which will permit separation of ions that enter the accelerator as hydrides.

As the first step in the construction of this beamline, the  $90^\circ$  electrostatic analyzer has been delivered to ANTARES and is undergoing testing. This unit was manufactured by DanFysik (Denmark) and has a mean radius of 2.5 m with a plate separation of 25 mm. The plates have a spherical geom-

etry and so the analyzer is double focusing for transmitted ions with a nominal maximum rigidity of  $E/q = 7.6$  MeV and it has an energy dispersion of 5000 mm in the image plane. The analyzer has been voltage conditioned in the factory beyond the rated voltage of  $\pm 80$  kV.

Initial testing of the ESA has commenced at ANTARES. For these tests, the ESA has been mounted on the  $0^\circ$  port of the high energy analyzing magnet and the focal and energy dispersive properties of the ESA have been assessed. Results of these beam tests will be presented at the conference. The results of additional tests with existing equipment aimed at assessing the performance of the ion source, terminal strippers and TOF system will also be presented.

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#### A LONG-TERM RECORD OF UPWELLING FROM THE SANTA BARBARA BASIN, SOUTHERN CALIFORNIA

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A record of radiocarbon ages of surface ocean water in the Santa Barbara Basin, southern California, has been derived from AMS  $^{14}\text{C}$  measurements on samples of the pelagic pteropod *Limacina helicina*, preserved in varved sediments. Ocean reservoir ages, derived from these data and the atmospheric (tree-ring)  $^{14}\text{C}$  record, vary from 600 to  $>1000$  yr, and show clear trends over time. The record suggests that increased rates of upward mixing of  $^{14}\text{C}$ -depleted subsurface water began *ca.* AD 1410 and again *ca.* AD 1660, and that these excursions are superimposed on a long-term trend towards decreased upwelling.

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#### A PROGRAM FOR LONG-TERM RETENTION STUDIES OF $^{14}\text{C}$ -LABELED COMPOUNDS IN HUMANS USING THE LUND AMS FACILITY

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Organic compounds labeled with  $^{14}\text{C}$  are routinely and to a wide extent used in clinical medicine and biomedical research to demonstrate abnormalities in the metabolism, study liver function or demonstrate abnormal activity of gastrointestinal bacteria. There are, however, considerable uncertainties in the current estimates of absorbed doses to man from  $^{14}\text{C}$ -labeled radiopharmaceuticals, mainly due to lack of reliable long-term biokinetic data the difficulty of performing highly sensitive measurements of the retention of  $^{14}\text{C}$  in the body by conventional radiometric techniques. In this paper



a project is described using AMS to perform investigations of the long-term biokinetics of  $^{14}\text{C}$ -labeled pharmaceuticals in humans. So far, the investigations have been performed by analysis of expired  $^{14}\text{CO}_2$ . The use of the AMS technique makes it possible to follow the turnover of the administered radionuclide for extensive periods and also to lower the doses administered.

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## INVESTIGATION OF THE ALUMINIUM BIOKINETICS IN RATS USING $^{26}\text{Al}$

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The radioisotope  $^{26}\text{Al}$  was used as a tracer to investigate the aluminium biokinetics in rats. Two studies were performed. In the first study, 20 ng  $^{26}\text{Al}$  were administered orally to 12 Sprague-Dawley rats. Six of them were controls. Renal failure was induced by 5/6 nephrectomy to another six animals. Samples of blood serum, urine, trabecular bone, liver and spleen were taken 24 h after application. The aim of this study was to investigate differences between healthy and nephrectomized animals in gastrointestinal resorption, compartmentalization and elimination. In the second study 0.2 ng  $^{26}\text{Al}$  were applied intravenously to 15 controls and 15 uremic rats. The rats were sacrificed at different times of 1, 5, 24, 125, 625 and 3000 h after administering the  $^{26}\text{Al}$ . Tissue samples of blood serum, urine, trabecular bone, liver, spleen and muscle were taken. The aim of this study was to measure  $^{26}\text{Al}$  in all important organs of the rats as function of time, to establish a compartment model simulating the aluminium kinetics in the body, and to assign the model compartments to organs and tissues. This model is to determine rate constants between organs or compartments.

An open compartment model consisting of a central compartment representing the blood serum and three peripheral compartments was developed which is able to describe the aluminium biokinetics in rats. Time curves of aluminium concentrations in rats cannot be obtained simply by scaling down the time curves of human subjects (Steinhausen *et al.*, in press). The compartment model was applied to the present studies and to Meirav *et al.* (1991), Sutton *et al.* (1991) and Jouhanneau *et al.* (in press). The model is able to describe the measurements. Time constants are derived.

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**AMS AT LOW ENERGIES***M. SUTER, ST. JACOB*

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The investment and the running costs of an AMS facility are related to the size of the installation. It is therefore worthwhile to study what the smallest possible size of a Tandem AMS system might be for reliable and efficient analysis of long-lived radionuclides in natural concentrations. Pioneering work in this field has been done by the IsoTrace group in Toronto (Lee *et al.* 1984). More recently, several concepts have been discussed for small  $^{14}\text{C}$  AMS systems for biomedical applications where radioisotope concentrations are significantly higher than in natural samples (Purser 1993).

This paper describes investigations made at the ETH/PSI AMS facility at low terminal voltages (*ca.* 1 MV). The goal was to obtain data for the design of a new generation of AMS facilities. In case of  $^{14}\text{C}$  it is known that its isobar  $^{14}\text{N}$  does not form negative ions. The main problem is the separation or destruction of molecules, such as carbonhydrides and  $^7\text{Li}_2$ .

Destruction cross sections for the molecules have been determined by studying the counting rates as a function of stripper density. We also performed tests with various detection and identification systems. Besides the traditional  $\Delta\text{E-E}$  gas counter arrangement, a combination of passive  $\Delta\text{E}$  (polypropylene foil of  $100\ \mu\text{g}/\text{cm}^2$ ) and a surface barrier detector placed 40 cm behind the foil were studied. The secondary electrons produced in the foil were detected in a microchannel plate and used for a time-of-flight measurement in combination with the surface barrier detector. Such a combination provides an excellent identification system at low energies. It allows separation of  $^{12}\text{C}$ ,  $^{13}\text{C}$  and  $^{14}\text{C}$  independently of their energy.

At 1 MV the stripping yield with Ar gas for charge state  $2^+$  was measured to be 56% which is comparable to the yield at 2.5 MeV for charge state  $3^+$ . Presently, the  $^{14}\text{C}$  background is a few times  $10^{-14}$  for samples processed with the conventional  $\text{CO}_2$  reduction technique using Co powder. With graphite the background is in the  $10^{-15}$  range. The remaining background seems partially to be caused by  $^7\text{Li}_2$ . Even when the molecules break up in the stripper, there is a chance that both fragments are in charge state  $1^+$  and reach the final detector almost simultaneously and thus mimic a  $^{14}\text{C}$  event. With an appropriate sample preparation this background should be reducible. Improvement of the detector system should also allow a finite separation of this Li background.

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**STATUS REPORT OF THE PSI/ETH AMS FACILITY**

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The PSI/ETH tandem accelerator laboratory has served as a multi-user multi-isotope AMS facility for >10 yr. Since 1982 *ca.* 35,000 individual samples have been analyzed for their  $^{10}\text{Be}/\text{Be}$ ,  $^{14}\text{C}/\text{C}$ ,  $^{26}\text{Al}/\text{Al}$ ,  $^{36}\text{Cl}/\text{Cl}$  or  $^{129}\text{I}/\text{I}$  isotopic ratios in routine AMS measurements. During the past, the throughput has increased constantly and >4000 samples have been analyzed per year in 1994 and 1995. In addition, a great variety of other long-lived radionuclides such as  $^{32}\text{Si}$ ,  $^{41}\text{Ca}$ ,  $^{59}\text{Ni}$ ,  $^{60}\text{Fe}$  and  $^{126}\text{Sn}$  have been analyzed using AMS detection techniques. In this contribution, we will discuss the technical and operational details of the AMS system and give an overview on new developments which have improved efficiency, sensitivity and accuracy of the measurements. The numerous applications connected with these measurements will not be covered.

The system is based on a 6 MV EN tandem accelerator. In the original set-up only one simple magnetic mass spectrometer has been used at the low energy side of the accelerator. A dedicated heavy ion spectrometer has been added for the injection of ion beams heavier than carbon. Pulsed beam operation is possible with both injection systems. They are equipped with our standard caesium sputter source utilizing a Cs gun to produce the caesium beam. A high current Cs sputter source is now also connected to the heavy ion injector. A new ion source with a micro-beam Cs gun is currently being added to the heavy ion injector. This system will be used for stable trace element analysis exploiting AMS detection techniques. At the high energy side of the accelerator only electrostatic beam optic elements are used to perform a mass independent beam transport. A high resolution mass spectrometer is used for the final separation.

Several techniques are applied to detect the analyzed beams, to identify radionuclides and to separate isobars. The standard technique uses a gas ionization detector to measure the specific energy loss of the ions stopped. We will discuss our experience and give details on isobar separation capability. A gas-filled magnet is available as an additional isobar separator. It is used for  $^{32}\text{Si}$ ,  $^{59}\text{Ni}$  and  $^{60}\text{Fe}$  detection in combination with a gas ionization detector. Due to the limited amount of beam energy available from an EN tandem, isobar separation is limited strongly for higher masses using the techniques described above. We have researched the detection of projectile X-rays (PXD) with respect to its isobaric separation potential. Examples are given and limitations are discussed. Time-of-flight systems can be used to improve stable isotope suppression and, if the start detector is used as a passive absorber, for isobar separation. The latter has potential for the detection of radionuclides at low energies. For radionuclides analyzed routinely, the measurement set-up is described and the operational conditions are summarized. In particular, we discuss performance parameters, detection limits, sources of background and accuracy for  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$  and  $^{129}\text{I}$  measurements.

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## RECENT APPLICATION OF $^{14}\text{C}$ MEASUREMENTS TO SOIL ORGANIC MATTER DYNAMICS

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Radiocarbon may be used in several ways to understand the soil carbon cycle: 1) the observed increase in  $^{14}\text{C}$  in soil organic matter since 1960 (due to the incorporation of carbon labeled with  $^{14}\text{C}$  created by atmospheric testing of nuclear weapons) indicates the amount of carbon turning over on decadal and shorter timescales in soils; 2)  $^{14}\text{C}$  measurements of fractionated organic matter may be used to identify extremely labile or refractory components of soil organic matter; and 3) the  $^{14}\text{C}$  content of decomposing organic matter (measured as the  $^{14}\text{C}$  content of soil  $\text{CO}_2$  or  $\text{CH}_4$ ) is a sensitive indicator of the turnover time of more rapidly cycling carbon. For the purpose of studying decadal changes in soil C and  $^{14}\text{C}$ , two components of soil organic matter are needed: fast cycling C (with turnover times of decades or less and consisting of undecomposed plant matter and exchangeable C on mineral surfaces) and passive C (stabilized for centuries or longer by mineral surfaces or aggregates). The relative amounts of fast cycling and passive C depend on soil forming factors, such as climate, vegetation, parent material and time. To study the influence of soil development and mineralogy on the amount and residence time of C in passive soil organic matter, we studied a chronosequence of 6 sites in Hawaii (300 y to 4,100 ky) with similar parent material, topography, vegetation, and climate. The amount and turnover time of organic matter in these soils is clearly associated with mineralogical changes in the soils over time. We studied the effect of climate on the turnover of fast cycling soil C using the observed  $^{14}\text{C}$  increase since 1960 in soils located along an elevation transect in the Sierra Nevada mountains, California. Average turnover times of fast cycling C in soils for which moisture is not a limiting factor are strongly related to temperature, ranging from <10 yr at 18°C (mean annual temperature) to 60–80 yr at 3°C. Comparison of the results from the Sierra Nevada with other sites shows a general relation between temperature and fast-cycling organic matter. An estimate of the amount of fast-cycling C globally, coupled with the strong dependence of C turnover on temperature for this fraction of soil organic matter, shows that transient C storage and loss in soils accompanying temperature anomalies may significantly affect atmospheric  $\text{CO}_2$  on interannual to decadal timescales.

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## THE AUSTRALIAN PROGRAM ON ACCELERATOR DATING OF ABORIGINAL ROCK ART

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A research program on AMS/ $^{14}\text{C}$  dating of aboriginal rock art has been funded by the Australian Research Council, the Australian Institute of Nuclear Science and Engineering and the Australian Institute for Aboriginal and Torres Island Studies. Present projects include detailed research into the probable antiquity of the rock art of Chillagoe and Laura, North Queensland, the Kimberly, Western Australia, and Olary District, South Australia and the Sydney Sandstone Basin in NSW. This archaeological program is encouraged and supported by the Aboriginal Tribal Councils representing the traditional owners of the Australian rock art sites. A variety of materials is being analyzed, including

pigments, oxalate minerals, silica coatings, plant fibers, carbonized plant matter, fatty acids, bees-wax and mud-wasp nests. Different sample processing techniques are being explored at Lucas Heights Research Laboratories, at the Australian National University, at Data-Roche Watchman Inc. and at Texas A&M University, including low-pressure plasma techniques and laser extraction methods. We recently used AMS/ $^{14}\text{C}$  to date a microstratigraphic sequence in 2 mm-thick rock surface accretions, a very useful method to study rock art concealed within the encrustation. Using dilution techniques developed at ANSTO, samples corresponding to 20  $\mu\text{g}$  of carbon have been analyzed. The significance of our first results will be discussed.

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## THE ANTARES AMS CENTRE: RECENT DEVELOPMENTS AND RESEARCH PROGRAM

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A comprehensive AMS facility has been developed during the past six years at the Lucas Heights Science and Technology Centre, where the long-lived radioisotopes  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{10}\text{Be}$  and  $^{129}\text{I}$  can be measured with high precision and throughput (Hotchkis *et al.*; Smith *et al.*; Hotchkis *et al.*, this conference; Fink *et al.*, this conference).

The key components characterizing the ANTARES AMS spectrometer are a 59-sample high intensity ion source, a fast isotope cycling system and two dedicated AMS beamlines. A gas stripper with terminal pumping and optical telemetry of gas pressure has been recently installed in addition to the foil stripper. Sample preparation laboratories are an integral part of the AMS facility (Jacobsen *et al.*, this conference). A considerable effort is presently devoted to support advanced research programs in Quaternary science and global climate change in collaboration with Australian research institutions and universities. This program is sponsored by the Australian Institute of Nuclear Science and Engineering, the Australian Research Council and the National Greenhouse Advisory Committee.

The paleoclimatic records of the Quaternary period contain evidence of widespread and dramatic changes that can be used to understand responses and processes in environmental systems on a global scale. Australia has a responsibility to contribute to these investigations because its area, surrounding oceans and Antarctica, represents a very significant part of the Southern Hemisphere. Corals, deep sea sediments, tree rings and ice cores are some of the outstanding archives present in the Australian region spanning a meridian from the Antarctic to the equator. There are a variety of environmental situations to which only Australian data can contribute (Ribbe *et al.* this conference; Murphy *et al.*, this conference; Levchenko *et al.*, this conference; Fink *et al.*, this conference; Barbetti *et*

*al.*, this conference). The records of the human presence in this region spans the last 40,000 yr and is directly related to environmental changes in the Pleistocene (Tuniz *et al.*, this conference).

Our environmental monitoring program for nuclear safeguards, sponsored by the Australian Safeguards Office and the IAEA, is based on the detection of  $^{129}\text{I}$  (and other long-lived radioisotopes produced by fission or neutron activation) to trace specific nuclear activities such as reactor operations, uranium enrichment and weapons testing. AMS measurement of actinides in environmental samples is also being developed within this program (Smith *et al.*, this conference).

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## IMPROVEMENTS OF THE AMS FACILITY AT UTRECHT

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Adaptations have been made to the Utrecht facility based on an EN tandem accelerator. Especially for  $^{14}\text{C}$  analyses, a stable and efficient stripping has been accomplished through the circulation system in the gas in stripper based on a 360 l/s turbopump. Rapid alternation of the isotope beams of Be, C, Al, and Cl is obtained with a new beam switching system. A preset sequence of HV-pulses can be chosen with amplitudes between  $-3$  and  $+6$  kV and pulse lengths between 40 and 1000  $\mu\text{s}$ . To improve the detection limit, especially for the heavier radionuclides, we have installed a  $90^\circ$  electrostatic analyzer prior to the existing  $90^\circ$  injection magnet. Both analyzers are matched to form an achromatic system with a mass resolution  $m/\Delta m = 200$ . At the high-energy end we replaced the  $90^\circ$  existing analyzing magnet by a larger ( $R = 110$  cm) and double focusing one. The computer-controlled system allows us to perform automated measurement of batches containing 22 samples. For  $^{14}\text{C}$ , the analytical precision reached is 0.3% for recent material, and the detection limit allows resolution of residual activities back to 50,000 yr. For the heavier radionuclides the analytical precision reached is 1%, and the detection limit is 10–14.

## AMS IN GRONINGEN: A SURVEY OF PROJECTS

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Since the Groningen AMS system became operational for  $^{14}\text{C}$  analysis (summer 1994), *ca.* 2000 samples have been analyzed for a broad range of applications and projects. Here we provide a survey of the results obtained thus far.

In peat-bog deposits, a high resolution study showed natural  $^{14}\text{C}$  variations, enabling wiggle matching to obtain more or less absolute dates. In peat bogs an unexpected reservoir effect is found, as well as indications for rapid climatic change. With AMS wiggle matching we have studied Holocene peat-bog deposits in the Netherlands, and varved sediments from sites in Poland and Japan.

A new application has been developed in soil sciences. A new Soil Organic Matter (SOM) fractionation method was developed recently and applied to both maize and pasture. The results indicate that fractions exist with ages ranging from 2 yr (coarse fractions) to >3000 yr (mineral associated with carbon). Maize cropping induced a strong rejuvenation of the finer fractions which represent colloidal organic material in association with soil particles. Because of the small sizes of the fractions, only AMS can be applied; this offers new possibilities for studying SOM dynamics in soils.

In hydrology, a large program has been started for isotopic research in Botswana, where for  $^{14}\text{C}$  amounts of water only possible with AMS are available. Together with stable isotopes ( $^2\text{H}$ ,  $^{13}\text{C}$ ,  $^{18}\text{O}$ ) unique information on water-resource development in the region will become available.

In oceanography, we participate in some WOCE activities. At present we have measured  $^{14}\text{C}$  extracted from ocean water profiles in the Philippine Sea. We have also constructed a special system for ocean-DOC analysis. The first experiments on the North Sea are presently performed, as well as some key locations in the oceans. The goal is a better understanding of the role of DOC as part of the global carbon cycle. A related project just started concerns origin and characterization of particulate organic matter (POC) in estuaries. Fractions are analyzed with Py-GC-MS for  $^{13}\text{C}$  and AMS for  $^{14}\text{C}$ .

Atmospheric research has been devoted mainly to a system for "event trapping" at Kollum, a station set up 30 km northwest of Groningen for off-line isotopic analysis of tropospheric  $\text{CO}_2$  ( $^{13}\text{C}$  and  $^{18}\text{O}$  by MS, and  $^{14}\text{C}$  by AMS).

## THE BIOSCIENCE AMS PROGRAM AT LLNL

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A prediction at AMS-5 expected that by the year 2000 there would be more AMS facilities involved in bioscience applications than in geoscience research. With more than half of that time now passed, AMS retains the ability to surprise biological scientists with the sensitivity for isotope tracing, and numerous interesting studies have now been performed. However, the enthusiasm has not translated into large programs at other than a few isolated sites. The Center for AMS continues a vigorous demonstration of the usefulness of AMS in studies of pharmaco-kinetics, environmental tracing, toxicology, molecular damage and repair, elemental distribution, and human health risks.  $^{14}\text{C}$  remains the primary isotope used in labeled organic compounds, but we have shown the sensitivity of  $^3\text{H}$  detection, opening pharmacokinetics to unprecedented double-labeled studies.  $^{41}\text{Ca}$  studies form the basis of another contribution submitted separately.  $^{59}\text{Ni}$  exploratory efforts have been performed with rats after  $^{63}\text{Ni}$  capability had been shown useful for neutron dose reconstruction reported separately. Initial efforts in developing  $^{79}\text{Se}$  for seleno-protein tracing are on hold while further spectrometer equipment is installed. The much-requested (by biomedical scientists) availability of  $^{129}\text{I}$  measurements for labeled proteins and enzymes awaits construction of a new heavy ion detection line and further research into rapid sample conversion techniques. Using radiocarbon,

extensive tracing of human carcinogens such as benzene and phenyl-imidazo-pyridine have shown that some organic compounds are truly linear in their dose extrapolation to low human exposures, while others increase in tissue availability at these low levels. Multi-sector tandem mass spectrometry has been combined with AMS to determine the actual molecular targets of human carcinogens. AMS immunoassays are being used to look at environmental contaminants such as atrazine and PCBs. Finally, new programs have begun between CAMS and a consortium of campuses that promise to bring a new spectrometer to LLNL for continued expansion of bioscience AMS at LLNL and the University of California.

Work was performed under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-ENG-48.

### **THE $^{36}\text{Cl}/^{36}\text{Cl}$ METHOD FOR DETERMINING EXPOSURE TIME AND EROSION RATES OF SURFACES**

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Chlorine-36 is produced by interaction of galactic cosmic rays with rocks on the surface of the Earth by two different types of nuclear reaction: 1) spallation of calcium and potassium produces  $^{36}\text{Cl}$  with an experimental decrease with depth; and 2) thermal and epithermal neutron activation of  $^{35}\text{Cl}$  produces  $^{36}\text{Cl}$  with a profile that first increases with depth, goes through a transition maximum at *ca.* 65 g/cm<sup>2</sup>, and then decreases exponentially below *ca.* 100 g/cm<sup>2</sup>. Because of this opposite dependence on depth near the surface, the ratio of production from spallation to that from neutron activation in a single surface sample is a sensitive measure of erosion. Production by spallation alone, as is also the case for  $^{10}\text{Be}$ , is an excellent means for determining exposure time if erosion can be neglected or corrected for.

Production systematics for  $^{35}\text{Cl}(n,\gamma)^{36}\text{Cl}$  in core and surface samples with negligible erosion have been studied in some detail (Dep *et al.* 1994a,b). Because of the variable and low chloride concentration in fluid inclusions of quartz, this approach has been discontinued. We are currently testing methods for determining both the spallation and neutron activation components of  $^{36}\text{Cl}$  using mineral fractions with low- and high-chloride concentrations. From these, we can determine the rock exposure age and erosion history, as long as the total erosion is under *ca.* 65 g/cm<sup>2</sup>. Results will be presented for bulk rock and mineral separates in surfaces and core samples from the top of Mt. Evans, elevation 4300 m.

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## AMS MEASUREMENTS OF THE $^{14}\text{C}$ DISTRIBUTION IN THE PACIFIC OCEAN

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Over the last 5 yr, the National Ocean Sciences AMS Facility at Woods Hole Oceanographic Institution has analyzed over 3700 seawater samples from World Ocean Circulation Experiment (WOCE) cruises<sup>1</sup> in the Pacific Ocean. The data set accumulated so far covers a major part of the ocean basin with emphasis on the upper water column (0–2000 m). Statistical aspects of the data analysis will be discussed as well as database related issues. We will present a three-dimensional model of the current  $^{14}\text{C}$  distribution in the region of coverage. Using this model, we will attempt to quantify the  $^{14}\text{C}$  transport in comparison with GEOSECS data from 1973.

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## PROGRESS OF THE NOSAMS FACILITY WOCE $^{14}\text{C}$ DATA ANALYSIS

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The NOSAMS facility has processed over 3700 WOCE<sup>1</sup> Pacific seawater samples so far. In comparison, during GEOSECS a total of 876 Pacific large volume samples were collected and analyzed. The 20-yr progression of the  $^{14}\text{C}$  distribution from GEOSECS to WOCE will be shown for overlapping stations. We will discuss the status of the operations and give a preview of the results to be presented at AMS-7.

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## EVOLUTION OF $\Delta^{14}\text{C}$ IN THE SURFACE NORTH ATLANTIC OCEAN

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Recent work using accelerator mass spectrometry (AMS) and micro-sampling methods has shown that the carbonate shell of the long-lived mollusc (*Bivalvia*) *Arctica islandica* can be used to reconstruct long-term, high-resolution  $\Delta^{14}\text{C}$  time series for the colder, higher-latitude northern North Atlantic Ocean (Weidman and Jones 1993). Here, time histories of the ocean's bomb- $^{14}\text{C}$  signal from three sites in the northeastern North Atlantic Ocean (southern North Sea, northern Norway, northern Iceland) are reported. These records, combined with previously published *Arctica islandica*-derived (Weidman and Jones 1993) and coral-derived  $\Delta^{14}\text{C}$  time histories (Druffel 1989), now permit a more complete overview of the evolution of the radiocarbon signal in surface North Atlantic Ocean than has been previously possible.

Pre-bomb (~1950) surface North Atlantic  $\Delta^{14}\text{C}$  values average  $-59\text{‰}$ , and are most depleted in the Labrador Sea (~  $-70\text{‰}$ ) and most enriched in the Sargasso Sea (~  $-50\text{‰}$ ). North Sea and Norway

pre-bomb signals now show large depletions (~30‰) in the 1950s relative to the 1940s, perhaps related to the Sues effect. However, the Iceland pre-bomb signal shows no depletion at all.

Initial appearance of the bomb-<sup>14</sup>C is synchronous in all North Atlantic  $\Delta^{14}\text{C}$  ca. 1959 ( $\pm 1$  yr). All North Atlantic  $\Delta^{14}\text{C}$  records show a steep rise during the early-to-mid 1960s and reach maxima in the late 1960s to late 1970s. The North Sea record has the largest bomb-signal amplitude (~320‰) and the Iceland record has the smallest (~120‰). The prolonged residence time of shallow shelf water is suspected to have caused the extremely large amplitude and advanced phase of the North Sea signal relative to their North Atlantic signals. The North Atlantic bomb-<sup>14</sup>C signals show a sharp contrast between regions influenced by the subtropical gyre (Florida, Bermuda, North Sea) with the former being more depleted. These results likely reflect the contrast between the depths of the winter mixed-layer in the northern and southern regions (Robinson *et al.* 1979), but are also consistent with a depleted or deepwater contribution to the subpolar gyre (Tsuchiya 1989). A small east-west difference in the relative enrichment of the Norwegian signal *versus* the Georges Bank and Iceland signals is also in agreement with deeper mixing in the western section of the subpolar gyre, and the poleward advection of surface Atlantic water into the Norwegian Sea.

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#### A THEORETICAL STUDY OF SOME NEGATIVE IONS OF INTEREST TO ACCELERATOR MASS SPECTROMETRY

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Recent experiments and theoretical calculations provide evidence that many atoms with closed subshells form stable negative ions. This was quite unexpected, since it had long been believed that these atoms, for example Ca, Sr and Ba, could not bind an extra electron to form stable negative ions. This view was abandoned with the prediction and observation of stable  $\text{Ca}^-$ ,  $\text{Sr}^-$ ,  $\text{Ba}^-$  and  $\text{Ra}^-$ .

The purpose of the present work is to study the stability of negative ions, many of which are of interest and importance for AMS. In particular, the effects of interactions between valence electrons and relativity on  $\text{Ca}^-$ ,  $\text{Ba}^-$ ,  $\text{Au}^-$ ,  $\text{Al}^-$ ,  $\text{Tl}^-$  and many other ions have been investigated.

#### THE GRONINGEN AMS TANDETRON

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The second machine of the so-called third generation AMS (employing simultaneous injection through a recombinator) is now in operation at Groningen for ca. 2 yr. The machine was built by HVEE in Amersfoort and is dedicated to <sup>14</sup>C analysis.

The accelerator is a tandemron operating at 2.5 MV and equipped with a 59-sample ion source. The full automation enables overnight operation on a routine basis. In a typical measurement batch, every fourth sample is a standard. We use sucrose (IAEA-C6) as an internal working standard.

Here we present a review of the first two years of operation, where >2000 measurements were performed. Overall performance per batch is better than 1‰ for  $\delta^{13}\text{C}$  and better than 0.5 pMC for  $^{14}\text{C}/^{12}\text{C}$ . Reproducibility tests indicate that the machine performance is better than these numbers. The final accuracy is mainly determined by target quality.

The pure machine background (*i.e.*, measured without target) is negligible (<100 ka). From background target tests, we conclude that for real measurements the background is *ca.* 50 ka.

### MOLECULAR BREAKUP OF $\text{CO}_2$ AND HOLD-UP TIME IN A MICROWAVE ION SOURCE

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The production of  $\text{C}^+$  in a microwave ion source injected with  $\text{CO}_2$  gas has been investigated. The microwave ion source was operated with pure  $\text{CO}_2$  feed gas and the extracted beam was magnetically analyzed. Efficient breakup of the  $\text{CO}_2$  molecule was observed. In a separate experiment, a continuous flow of argon maintained the discharge and  $\text{CO}_2$  gas was pulsed into the source through an electromagnetic valve closely coupled to the plasma chamber. The  $\text{C}^+$  and  $\text{O}^+$  components of the beam fell to one-half their original intensity <20 sec after the electromagnetic valve was closed. These results are considered promising for such applications as accelerator mass spectrometry of C isotopes in gaseous samples and for ion sources for radioactive ion beam facilities.

### $^{14}\text{C}$ STUDIES ON THE GISP-2 ICE CORE: DO POLAR ICE SHEETS CONTAIN A RECORD OF PAST GALACTIC COSMIC RAY FLUX?

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Recent developments have made it possible to accurately measure the very small amounts of  $^{14}\text{CO}_2$  present in samples of glacial ice (Wilson and Donahue 1990). This  $^{14}\text{CO}_2$  may originate as atmospheric  $^{14}\text{CO}_2$  trapped in voids in the ice or may be produced *in-situ* by cosmic ray spallation of oxygen atoms in the ice (Fireman and Norris 1982). In this paper we show how the amount of  $^{14}\text{CO}_2$  present in glacial ice is related to many factors, including the age of the sample, rate of cosmogenic  $^{14}\text{C}$  production and the degree of recrystallization of the snow-pack during firnification. The  $^{14}\text{CO}_2$  concentrations from the GISP-2 ice core for the last 40 ka will be presented. Two important cases in which simplifying assumptions can be made about many of these factors will be discussed. Within the Holocene portion of the GISP-2 core,  $^{14}\text{C}$  dating can be used to estimate the age of ice samples. Within the section from the last glacial period,  $^{14}\text{CO}_2$  measurements on samples of the same core were used to estimate cosmogenic  $^{14}\text{C}$  production and thus the galactic cosmic ray flux. Measurements of cosmogenic  $^{14}\text{C}$  production rate for the period 40–17 ka suggest that the galactic cosmic ray flux remained constant at current levels for the period 30–17 ka after a perturbation, possibly a

supernova event, which started *ca.* 35 ka ago and ended *ca.* 29 ka ago. Before this perturbation the data suggests that the galactic cosmic ray flux was 2.7 times greater than at present.

### TRACE-ELEMENT ANALYSIS OF MINERAL GRAINS USING ACCELERATOR MASS SPECTROMETRY – FROM SAMPLING TO INTERPRETATION

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AMS of trace elements in natural materials has been limited to a few labs around the world, the majority of extant data stemming from work in Rochester, Toronto (*e.g.*, Kilius *et al.* 1984) and Oxford. Early analyses utilized time-of-flight systems, whereas recent studies at IsoTrace employ high-resolution magnetic and electric analyzers. Most data concern rare precious metals: Au, Ag, Re and the six platinum group elements (PGE). Most measurements are at ppb to ppm levels, observed abundances ranging from ~0.01 ppb to 0.1 wt.% (Wilson *et al.* 1995). Many public-domain results may be invisible to the broader AMS community; a partial bibliography of PGE-Au-Ag work lists 58 articles, abstracts, reports and theses (Wilson 1994: 33–37).

The key feature of trace-element work at IsoTrace in the 1990s has been the use of AMS results as one component in integrated studies of minerals and metals in rock, meteorite and archaeological samples (*e.g.*, Pavlish *et al.* 1995). Major and minor elements in chosen grains are often determined by electron microprobe analysis, and trace elements may be analyzed by proton microprobe (PIXE) or SIMS, prior to (more) destructive AMS analysis using a Cs<sup>+</sup> primary ion beam 300–1000  $\mu\text{m}$  in diameter. Until now, *in-situ* analysis has been limited to conductive materials which, coupled with the relatively poor spatial resolution, has limited user interest. Reliable interpretation of the results must be based upon a comparison of all available data: textures observed by reflected-light microscopy/SEM, plus the *in-situ* chemical data achieved on scales between 1 and 1000  $\mu\text{m}$ . It is suggested that one reason for the limited application of AMS in this context during the past 15 yr is the complex nature of the targets. Whereas homogenized (*e.g.*, Rucklidge *et al.* 1992) or preconcentrated samples can be treated as identical point targets, natural targets require many decisions concerning target selection.

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**TEXTURAL AND *IN-SITU* ANALYTICAL CONSTRAINTS ON THE PROVENANCE OF SMELTED AND NATIVE ARCHAEOLOGICAL COPPER IN THE GREAT LAKES REGION OF EASTERN NORTH AMERICA**

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*In-situ* ( $\mu\text{g}$ -mg,  $\sim 1$  mm) AMS analysis, as practiced with high-current Cs<sup>+</sup> ion sources, permits the characterization of small crystals and sherds of rare or valuable samples. These include small slivers of metallic articles, such as copper or brass, snipped from inconspicuous loci on archaeological metal artifacts. Samples from a project in which instrumental neutron activation (INAA) of  $\sim 200$ -mg aliquots is the standard bulk analytical method (Hancock *et al.* 1991), were re-examined using a combination of wavelength-dispersive electron microprobe (10-element WDS-EPM) and AMS methods. Metal shavings were mounted in two polished formats: 1) thin sections with 20 2–5-mm slivers, suitable for metallographic study and EPM, and 2) sets of 12 sherds and standards in thick circular mounts, also suited to AMS of precious metals (Wilson *et al.* 1994). The samples are metal artifacts from the Great Lakes region, principally Ontario and Minnesota, plus native Cu (mineral specimens) from the Keweenaw Peninsula of northern Michigan. AMS <sup>14</sup>C dating (Beukens *et al.* 1992) indicates that some projectile points date to as old as 7,000 yr BP. Examination of 11 coppers suggested a rapid optical method of distinguishing artifacts made from native Cu *versus* smelted (trade or kettle) copper imported from Europe. Five samples of native copper were visually homogeneous and free of inclusions. The six “smelter” coppers contain high but variable numbers of spheroidal cuprite (Cu<sub>2</sub>O) blebs, generally 1–10  $\mu\text{m}$  in diameter, indicative of incomplete separation of oxide slag from copper metal. Similar features were encountered in a later survey of 68 mm-scale slivers of other artifacts. Practical minimum detection limits (MDL) for the EPM are  $\sim 200$ –1000 ppm. This is far higher than ppb (PGE, Au) to ppm (Au, Ag) levels found for trace elements by AMS, so the data obtained are complementary but not directly comparable. EPM data are especially useful for microchemical typing (Zn, Sn, As) of associated brass trade items. The “sourcing” of coppers was confirmed by PGE-Au-Ag data obtained by AMS on 9 samples. Smelted Cu has higher Ag ( $\pm$ Pt) and much higher Au than native Cu. This is consistent with INAA data, which show that a suite of trace elements (Au, Ag, Sb, As) can reliably separate the copper types.

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**PRECIOUS METAL ABUNDANCES IN SELECTED IRON METEORITES: *IN-SITU* AMS MEASUREMENTS OF THE SIX PLATINUM-GROUP ELEMENTS PLUS GOLD**

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The metal phases of the coarser iron and stony-iron meteorites are well-suited to *in-situ*, single-crystal analysis by AMS. In contrast, the fine-grained Ni-Fe alloys of the Ni-rich ataxite (type IV) irons appear as micron-scale eutectoid intergrowths, broadly reminiscent of the quenched groundmass of NiS fire-assay beads, and in these samples AMS can only provide a mean analysis of many thousands of crystallites (nevertheless a small sample population relative to 100 to 1000-mg bulk analyses, which are several orders of magnitude larger still). The scale of sampling is especially relevant to *in-situ* ratio measurements of stable isotopes (Ding 1996; *see also* Ding *et al.*, this conference). The elevated levels of PGE and Au in bulk samples of iron meteorites render detection of these 7 precious metals very straightforward, despite the widely variable sensitivity of AMS to each element (Wilson *et al.* 1995). Detection is harder for Ag (very low abundance) and Re (very low negative ion yield). Six irons of types I, II, III and IV were studied at length: Cañon Diablo, Negrillos, Welland, Manitouwabing, Weaver Mountains and Hoba. A detailed treatment of the results is available (Wilson *et al.* 1993).

AMS data on plessite (fine-grained material in the IVs), kamacite and taenite suggest a variation in overall precious-metal abundances of a factor of 16 between the most-enriched iron (Negrillos, ΣPGE+Au = 270 ppm) and the least-enriched (Welland, 16–19 ppm). The use of chondrite-normalized PGE patterns is common in terrestrial petrogenetic and Ni-Cu-PGE mineral deposit studies. They are seldom used for classification of iron meteorites because of wide variations in pattern within certain classes. Yet the PGE “fingerprint” may still be valuable for well-constrained problems. The use of PGE patterns for a provenance study (“pairing”, in this case) of meteoritic iron masses is presented for Welland (IIIA), an 1888 find from southern Ontario. Few published data are available for Welland: comparison of a type sample with a smaller piece of unknown metal, with respect to PGE pattern, major-element chemistry (electron microprobe data) and texture, indicate strongly that the latter is a fragment of the same iron.

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## AN AUTOMATIC CYCLING SYSTEM FOR MEASUREMENTS OF ISOTOPE RATIO AT CIAE AMS FACILITY

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An automatic cycling system for isotope ratio measurement has been developed at the China Institute of Atomic Energy (CIAE) AMS facility. The main modification was focused on fast cycling injection of stable isotopes and radioisotopes of interest on the injector alternatively, and measurements of the stable isotopes behind the analyzing magnet. A new injector vacuum chamber insulated from the deflection magnet with two gap lenses has been constructed for beam energy modulation. The rebuilt analyzing magnet chamber and off-axis Faraday cups have been tested, the off-axis cups are movable for collection of different stable isotope currents, such as  $^9\text{Be}$ ,  $^{13}\text{C}$ ,  $^{27}\text{Al}$  and  $^{35}\text{Cl}$  ions while measuring  $^{10}\text{Be}$ ,  $^{12}\text{C}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$ , respectively. A micro-processor system has been used as a sequencer and timer to trigger the pulse power supply, to control the current integrator, the ion detector and computer data acquisition system. The pulse power supply is matched with injection of stable isotopes while the original power supply is held to inject radioisotopes. The pulse period and width are controlled by the micro-processor system, ranging from 0.1 to 9.9 s and from 1 to 99 ms, respectively.

## LEAVES TEACH US AIR POLLUTION

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Exhaust fumes from automobile engines are an important origin of air pollution. The contribution of exhaust fumes to air pollution was investigated by a simple method in which  $\delta^{14}\text{C}$  values in leaves were measured with AMS at MALT (The University of Tokyo).

At present, almost all engines use fossil fuel that is made of dead carbon. The  $^{14}\text{C}$  content in the air polluted by the exhaust fumes is expected to be low because of dilution by the dead carbon. Averaged  $^{14}\text{C}$  content in the air as  $\text{CO}_2$  gas is also expected to vary in different districts where the air pollution level is different. Plants grown in different districts have different  $^{14}\text{C}$  contents in their stems than in the leaves.

To investigate directly the  $\delta^{14}\text{C}$  values in  $\text{CO}_2$  gas is not easy because the  $\delta^{14}\text{C}$  in the air samples, which are usually collected in a very short time, are affected by many conditions, such as the season, temperature, wind, weather, *etc.* This is not the case for leaves, which live a comparatively long time.

The specimens used for the investigation are pine needles, which can be collected in any place in Japan. Similar species can also be collected on high mountains where the air pollution level is expected to be very low. The samples were collected in urban districts (Tokyo and Beijing) and, for comparison, collected in the suburbs of Tokyo and on a high mountain (3000-m altitude) in central Japan.

The samples were treated with AAA treatment and the resulting cellulose components were pyrolyzed to elemental carbon at  $450^\circ\text{C}$  in vacuum.  $^{14}\text{C}/^{12}\text{C}$  values in the samples were then determined by AMS.

The preliminary results reveal that the  $\delta^{14}\text{C}$  values of the urban leaves are lower than those of the leaves from a high mountain, which shows the apparent air pollution in the urban districts by combustion of fossil fuel.

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## <sup>26</sup>Al UPTAKE AND ACCUMULATION IN THE BRAIN

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We have demonstrated aluminum (Al) accumulation in the brain cell nuclei from patients with Alzheimer's Disease (senile dementia) using microbe particle-induced x-ray emission (PIXE) analysis (Yomoto *et al.* 1995a), and secondary ion mass spectrometry (SIMS) analysis (Yomoto *et al.* 1995b). Al is a highly neurotoxic substance and dialysis dementia in patients on hemodialysis is caused by contamination of dialysis fluid with Al. In this study, <sup>26</sup>Al accelerator mass spectrometry (AMS) was applied for the assays of Al uptake and accumulation in the brains of rats to investigate the cause of Alzheimer's Disease. When <sup>26</sup>Al was intraperitoneally injected into healthy rats, *ca.* 0.002% of the injected Al was incorporated into the brain (cerebrum) 5–270 days after single injection. <sup>26</sup>Al concentrations in the blood decreased remarkably after 75–270 days following the injection. As much as 17% of the <sup>26</sup>Al incorporated into the cerebrum was measured in the brain cell nuclei. The present study strongly supports the theory that Alzheimer's Disease is caused by irreversible accumulation of Al in the brain, and brain cell nuclei (Yomoto *et al.* 1992, 1993, 1995a,b; Kobayashi *et al.* 1990).

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**$^{26}\text{Al}$  AND  $^{67}\text{Ga}$  METABOLISM IN RATS**

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Because of lack of a suitable isotope and a sensitive technique of analysis, aluminum has been studied indirectly using analogues such as  $^{67}\text{Ga}$  ( $t_{1/2} = 78$  hr). Recently with the development of Accelerator Mass Spectrometry (AMS), it has become possible to use the artificially produced radionuclide of aluminum,  $^{26}\text{Al}$ , ( $t_{1/2} = 7.16 \times 10^5$  yr). AMS is used for measuring long lived and stable isotopes with the sensitivity of an attomole (10–18 mol). To compare aluminum and gallium metabolism,  $^{26}\text{AlCl}_3$  and  $^{67}\text{GaCl}_3$  were coadministered to rats intraperitoneally (IP) by injection and orally by gavage ( $n=3/\text{group}$ ). The injections contained 11 pCi  $^{26}\text{Al}$  and 30 uCi  $^{67}\text{Ga}$ . The oral dose contained 330 pCi  $^{26}\text{Al}$  and 50 uCi  $^{67}\text{Ga}$ . Blood was collected periodically and on day 8, following perfusion, blood, liver, kidney, femur, brain, pancreas, and spleen were collected and analyzed for  $^{67}\text{Ga}$  and  $^{26}\text{Al}$ . Of all the tissues studied,  $^{26}\text{Al}$  accumulation was greatest in the bone. Both  $^{67}\text{Ga}$  and  $^{26}\text{Al}$  accumulated in tissues as bone>spleen>kidney~liver>pancreas>brain.

Although the retention of  $^{26}\text{Al}$  was 2–3 orders of magnitude less than the retention of  $^{67}\text{Ga}$ , absorption and retention of  $^{26}\text{Al}$  (0.81%) was significantly less than  $^{67}\text{Ga}$  (2.22%). Intestinal absorption was determined by calculating the ratio of the  $^{26}\text{Al}$  retained in the tissues of the oral group and the  $^{26}\text{Al}$  retained in the tissues of the IP group.  $^{26}\text{Al}$  cleared from the blood faster and more completely than  $^{67}\text{Ga}$  in both groups even after accounting for the decreased intestinal absorption in the oral group. Since  $^{67}\text{Ga}$  does not mimic aluminum metabolism, AMS is the only technique available for tracer aluminum study.

**THE  $^{41}\text{Ca}$  BOMB PULSE AND ATMOSPHERIC TRANSPORT OF RADIONUCLIDES**L. ZERLE,<sup>1</sup> Th. FAESTERMANN,<sup>1</sup> G. KORSCHINEK,<sup>1</sup> K. KNIE,<sup>1</sup> J. BEER<sup>2</sup> and E. NOLTE<sup>1</sup>

Radionuclides produced as a by-product of nuclear weapons tests are very suitable tracers to study transport processes in the atmosphere or the ocean. Their input is sharp in time and bomb-produced concentrations are typically several orders of magnitude larger than the natural levels. The most studied bomb-produced radioactivities are  $^{90}\text{Sr}$  (UNSCEAR 1982; Joseph *et al.* 1971),  $^{14}\text{C}$  (Nydal 1968; Levin, Münnich and Weiss 1980)  $^{36}\text{Cl}$  (Elmore *et al.* 1982; Synal *et al.* 1990) and  $^{123}\text{Cs}$ .

For the first time, the  $^{41}\text{Ca}$  bomb pulse has been measured.  $^{41}\text{Ca}$  concentrations have been determined in Alpine ice of the Fiescherhorn glacier in the Aletsch region (44°N, 15°E, Switzerland) with accelerator mass spectrometry (AMS) with the Munich MP Tandem accelerator. The ice core was drilled at an altitude of 3750 m. After addition of 10 mg Ca carrier and chemical treatment,  $\text{CaH}_2$  samples were obtained for the ion source. Negative  $\text{CaH}_3^-$  ions were used for injection into the Tandem in order to suppress the disturbing isobar  $^{41}\text{K}$ . The AMS measurements were performed with the Wien velocity filter, a time-of-flight line and the gas-filled Q3D magnet spectrograph (Korschinek 1994). The detection limit for the  $^{41}\text{Ca}/\text{Ca}$  was  $10^{-15}$ . The  $^{41}\text{Ca}$  peak concentrations in the ice core have been observed to be *ca.*  $3 \times 10^6$  atoms of  $^{41}\text{Ca}$  per kg in the 1950s. From the time of occurrence of the peak, it has been found that  $^{41}\text{Ca}$  is produced essentially by the atoll bombs.

A universal box model able to describe atmospheric transport of radionuclides which are gaseous or attached to aerosols has been developed. The model has been applied to calculate the bomb pulses of  $^{14}\text{C}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  and measured the concentrations of  $^{14}\text{C}$  and  $^{90}\text{Sr}$  in the atmosphere. For the transport of radionuclides which are attached to aerosols as  $^{41}\text{Ca}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , sedimentation (gravitational settling) in the upper stratosphere had to be taken into account. It has been found that  $^{36}\text{Cl}$  is essentially produced by the explosions on barges, that bomb produced  $^{36}\text{Cl}$  is gaseous in the stratosphere and that its deposition on the Earth's surface is delayed to that of  $^{90}\text{Sr}$  by about one year. Time constraints for exchange processes between upper and lower stratosphere and troposphere and between northern and southern hemisphere and for gravitational settling in the upper stratosphere were obtained as well as neutron yields per Mt fusion and per Mt fission.

For cosmogenic atmospheric radionuclides, e.g.,  $^{10}\text{Be}$  and  $^{36}\text{Cl}$ , the model was used to calculate delays between solar activity modulated production and deposition in natural archives as Greenland or Antarctic ice.

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#### DISCUSSION ON THE RELIABILITY OF $^{14}\text{C}$ DATING OF POLLEN CONCENTRATES FROM LOESS-PALEOSOL SEQUENCES BY ACCELERATOR MASS SPECTROMETRY

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Dating pollen concentrated from paleosol samples is a way to improve the chronology of loess-paleosol sequences. Pollen generally consists of two fractions: 1) pollen deposited simultaneously with sediments can provide reliable data and 2) reworked pollen including older pollen before sediments deposition *in situ* and younger pollen preserved in a stable environment can provide reliable age, such as pollen from either a closed lake or paleosol, while pollen preserved in an unstable environment cannot provide reliable data (for example, pollen in eolian sand or loess in an area where winter monsoon is predominant and strong). It is suggested that the reliability of pollen dating can be evaluated by comparison with wood cellulose or charcoal ages from the same stratigraphic level.

The purpose of the present work is to study the stability of negative ions, many of which are of interest and importance for AMS. In particular, the effects of interactions between valence electrons and relativity on  $\text{Ca}^-$ ,  $\text{Ba}^-$ ,  $\text{Cs}^-$ ,  $\text{Au}^-$ ,  $\text{Al}^-$ ,  $\text{Tl}^-$  and many other ions have been investigated.

## CLIMATIC TELECONNECTION BETWEEN EAST ASIA AND THE NORWEGIAN SEA DURING THE LATE GLACIATION

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Episodes of high frequency SST changes in the Norwegian Sea during the late glaciation might be linked to meltwater forcing creating a large degree of instability. Four major climatic events have been well documented in diatom records from the Southeast Norwegian Sea. Here, we present proxy results from loess-paleosol and lacustrine profiles in the desert/loess transitional belt. Accelerator mass spectrometry age control of the profiles enables us to correlate the proxy data in detail with oceanic records. The results indicate that the East Asian monsoon climate experienced Bølling (13–12.5 ka BP), Older Dryas (12.5–11.75 ka BP), Allerød (11.75–11.2 ka BP) and Younger Dryas (11.2–10.0 ka BP) periods and was characterized by significant variability of precipitation. The variability of precipitation in East Asia is greater than that of SST in the Norwegian Sea. It is suggested that monsoon front precipitation produced by the interaction between cold air mass from polar and high latitude areas and warm moist air mass from ocean may fluctuate in response to high northern latitude temperature and pressure changes that influence the land-to-ocean pressure gradient.

## NEW INTERPRETATION OF THE $^{10}\text{Be}$ AND $^{26}\text{Al}$ CONTENT IN COSMIC SPHERULES

U. ZOPPI,<sup>1</sup> H. MATSUZAKI,<sup>1</sup> K. KOBAYASHI,<sup>1</sup> M. IMAMURA<sup>2</sup> and H. NAGAI<sup>3</sup>

The observed concentrations of cosmogenic nuclides in extraterrestrial matter are usually interpreted in terms of the exposure history of the irradiated specimen or in terms of the history of the cosmic radiation itself. For small meteorites in the microgram mass range we present a theoretical approach which allows a new interpretation of the measured concentrations. In particular, we show how to determine the size distribution of the interplanetary dust and how to investigate the related phenomena (radiation pressure, mutual collisions, atmospheric entry). For this, the concentrations of the radioisotopes contained in spherules from deep sea sediments were measured by means of the AMS technique. Due to the small sample size, the carrier amount was successfully kept very low—ca. 50  $\mu\text{g}$  for Be and between 100 and 200  $\mu\text{g}$  for Al.

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## DEVELOPMENT PLANS FOR THE AMS FACILITY AT THE INSTITUTE OF GEOLOGICAL AND NUCLEAR SCIENCES, NEW ZEALAND

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In this paper, we outline proposed upgrades to the New Zealand AMS facility for the next three years. Since the AMS facility began operating in 1987, ca. 10,000 graphite and 200 beryllium samples have been measured for a wide range of applications, mainly in archaeology and earth sciences. Demands for higher sample throughput, a wider range of cosmogenic isotopes, smaller sample sizes

and improved accuracy and precision require constant improvements to instrumentation and techniques. Proposed improvements include a new ion source, Pelletron charging system, gas stripping with terminal pumping, redesigned injection optics and a more sophisticated detector to improve sensitivity for rare cosmogenic isotope measurements. An integrated accelerator control and data acquisition system is under development, and a prototype of the data acquisition section has already been implemented.

*[Addendum]*

### **THE CHALK RIVER AMS SAMPLE CHANGER AND ION SOURCE**

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Most AMS ion sources share the need for a sample changer that reliably exchanges samples with good positional repeatability, incorporates a large magazine that can be easily loaded or exchanged and minimizes the possibility of sample-to-sample contamination. A new sample magazine, sample changer and ion source is in use at Chalk River that meets these criteria. It features a readily-accessible magazine at ground potential that is external to the ion source and high-voltage cage. The samples are held in an inert atmosphere and can be individually examined or removed, or exchanged *en masse* as a complete magazine without interruption of AMS measurements. Since only one sample is in the source at any time, sample-to-sample contamination is avoided. Sample changing is done pneumatically with a rabbit transfer system and two stages of differential pumping. It requires a few seconds for completion and can be performed without altering ion source potentials. At Chalk River this is routinely performed across a 200 kV potential. Sample positioning is precise, and hundreds of  $^{36}\text{Cl}$  and  $^{129}\text{I}$  samples have been measured over a period of several days without interruption or alteration of ion-source operating conditions.

## OVERVIEW OF THE WORKSHOP ON SECULAR VARIATIONS IN PRODUCTION RATES OF COSMOGENIC NUCLIDES ON EARTH<sup>1</sup>

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### INTRODUCTION

Measurements of cosmogenic nuclides made *in situ* in the Earth's surface are being used to help resolve a wide range of geologic and chronologic questions. Cosmogenic nuclides (<sup>3</sup>He, <sup>10</sup>Be, <sup>14</sup>C, <sup>21</sup>Ne, <sup>26</sup>Al, <sup>36</sup>Cl are presently used) can reveal rock exposure history information leading to estimates of timing of surface forming events, rates and styles of erosion, and timing and durations of episodes of burial. Depending on the problems being tackled, a significant source of error ( $\pm 10$ –25%) for any cosmogenic nuclide method is the present uncertainty in the spatial and temporal variability of the rates of production of these *in-situ* nuclides.

The Workshop on the Secular Variations in the Rates of Production of Cosmogenic Nuclides on Earth was hosted by Los Alamos National Laboratory on 2–5 February 1996 in Santa Fe, New Mexico. The workshop was a multi-disciplinary, international meeting of 38 scientists from seven countries (see below). The workshop gathered together not only geologists, chemists and physicists directly involved in the determination of *in-situ* production rates, but also workers in the related fields of paleomagnetism, nuclear cross sections, cosmic-ray transport and other branches of geochronology. This is a cross section of scientists that would never be found at any single societal meeting.

### List of Attendees

Bierman, P.	University of Vermont
Brook, E.	University of Rhode Island
Brown, E.	CNRS-IN2P3, France; University of Minnesota
Caffee, M.	Lawrence Livermore National Laboratory
Clapp, E.	University of Vermont
Clark, D.	University of Washington
Damon, P.	University of Arizona
Dunne, A.	PRIME Lab, Purdue University
Evenson, E.	Lehigh University
Finkel, R.	Lawrence Livermore National Laboratory
Flinsch, M.	New Mexico Tech
Gillespie, A.	University of Washington
Gosse, J.	Los Alamos National Laboratory
Graham, I.	Institute of Geological and Nuclear Sciences, NZ
Graf, Th.	University of California, San Diego; ETH Zürich
Harrington, C.	Los Alamos National Laboratory
Heisinger, B.	Technical University of Munich

<sup>1</sup>Note: Unless otherwise noted, cited references in all workshop abstracts can be found in the "Bibliography for *In-Situ* Production Rates" following this Overview.

<sup>2</sup>Geology and Geochemistry Group, EES-1, MS D462

<sup>3</sup>Astrophysics and Radiation Measurements Group, NIS-2, MS D436

<sup>4</sup>Geology and Geochemistry Group, EES-1, MS D462

<sup>5</sup>Chemical Science and Technology, CST-7, MS J514

Imamura, M.	Institute for Nuclear Study, University of Tokyo
Ivy-Ochs, S.	ETH Hönggerberg, Zürich
Jull, T.	University of Arizona
Klein, J.	University of Pennsylvania
Liu, B.	Los Alamos National Laboratory
Marti, K.	University of California, San Diego
Masarik, J.	Max-Planck Institute, Germany
McHargue, L.	University of Arizona
Nishiizumi, K.	University of California, Berkeley
O'Brien, K.	Northern Arizona University
Phillips, F.	New Mexico Tech
Phillips, W.	Los Alamos National Laboratory
Poeths, J.	Los Alamos National Laboratory
Reedy, R.	Los Alamos National Laboratory
Stamatakis, J.	South West Research Institute, Texas
Sternberg, R.	Franklin and Marshall College
Stone, J.	Australian National University
Swanson, T.	University of Washington
Stuiver, M.	University of Washington
Verosub, K.	University of California, Davis
Zreda, M.	University of Arizona

The group recognized the limits and various sources of systematic uncertainties of the cosmogenic nuclides methods, especially the many ancillary data sets used, such as those used in adjustments for elevation and geomagnetic effects. There was a unique opportunity for open discussion and evaluation of the geological and chronological assumptions for the major calibration sites. Input from the fields of paleomagnetism, radiocarbon dating and cosmic-ray physics helped to address questions as they arose.

### Workshop Goals

The following goals and questions were addressed during the workshop:

1. To discuss the measurement of *in-situ* cosmogenic nuclide production rates. *How accurate are the current measurements? How accurate do we need them to be and what are the limitations? How well do we know the relative production rates of cosmogenic nuclides? What should be involved in selection of calibration sites? What factors are important to consider? What are some suggestions for good calibration sites? What are the caveats involved in measuring production rates at calibration sites and what experiences can be shared to decrease the probability of repeating mistakes?*
2. To discuss models relating cosmic-ray intensity and nuclear cross sections to production rates. *What do the models tell us about spatial variability? What can they tell us about temporal variations? How good are the models? For example, if we accurately knew the Earth's magnetic dipole paleointensity, how accurately can we now calculate production rates on Earth? What are the uncertainties in the models and their magnitudes? Are these models worth pursuing? What is needed to improve the models? How strongly are cross section measurements needed?*
3. To summarize the current state of knowledge concerning temporal and spatial variability of production rates. *How well do we know the spatial and temporal dependencies? What literature exists? What needs to be done to improve our knowledge of the dependencies on altitude and latitude? Where should measurements of *in-situ* production rates be focused? What is the need for very low and very high latitude measurements? Are there problematic results published?*

4. To discuss surrogate records of changes in parameters that affect production rates. *What records can be used now? How good are they? What improvements are being attempted that will have an impact on production rate modeling within the next half decade? What models exist or are needed to calculate changes in production rates based on changes in such factors as the strength of the Earth's magnetic field? What records of prehistoric solar activity are available and how good are they?*
5. To make suggestions that will help direct future efforts to calibrate cosmogenic nuclide surface exposure dating methods.
6. To compile a bibliography of current relevant publications (see below).

### State of Production Rate Knowledge

The talks and discussions covered a wide range of topics and data. The reader is referred to the abstracts and bibliography to attain the most recent estimates of production rates. Estimates of the *in-situ* production rates of the cosmogenic nuclides currently used in exposure history applications were presented and evaluated at the workshop. Production rate estimates have been determined from measurements of the nuclides in geologic samples and artificial samples (such as water) and through numerical simulations of the reactions involved. It is clear that the uncertainty of a production rate for a given sample site is improving with the increasing number of empirically and experimentally derived production rates.

However, some important discrepancies are evident in the existing production rate estimates. The reasons for the discrepancies are complicated because multiple variables are involved, including 1) the calibration sites may have had uncertain exposure histories, *e.g.*, some sites may have been partially buried by till, snow, or ejecta; 2) there are inherent uncertainties in the calibration age for the site, due to either assumptions associated with the calibration age or the geomorphic relationship between the site for the calibration age and that for the cosmogenic nuclide sample (*cf.* abstract of Clark *et al.*); 3) the calibrations were done for different exposure durations and geographic locations—variations in the Earth's geomagnetic field strength with time and location can account for significant variability on a 20 kyr timescale and; 4) the uncertainties associated with the scaling that is used to normalize production rates to sea level and high latitude can cause additional deviations. It was generally agreed that multiple sites of calibration are needed to provide information and constraints on elevation, latitude and temporal controls on production rates. The need for cross calibration of the production rates of different cosmogenic nuclides was also emphasized.

New and previous empirical data, used to calculate or verify production rates for  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in quartz, were presented by Nishiizumi *et al.*, Bierman *et al.*, Gosse and Klein, Ivy-Ochs *et al.*, Brown *et al.* and Brook *et al.* The rates normalized for production at sea level and high latitude are in fairly close agreement (all were within  $5.6 \pm 0.5 \text{ atoms} \cdot \text{g}^{-1} \cdot \text{yr}^{-1}$ ) for times ranging from the present to >10 Myr.

The rates for *in-situ*  $^{14}\text{C}$  in quartz were reported by Jull and McHargue. The use of *in-situ*  $^{14}\text{C}$  work is relatively recent and more production rate measurements are immediately needed. Considering the importance of  $^{14}\text{C}$  in erosion rate and burial studies due to its short half-life, production rate refinement for this nuclide should get considerably more attention.

Data useful in constraining production rates of the cosmogenic noble gases  $^3\text{He}$  and  $^{21}\text{Ne}$  were presented by Brook *et al.*, Brown *et al.*, Poths *et al.*, Phillips, W. *et al.* and Marti and Graf. The current production rate estimates of these nuclides are in reasonable agreement for times >10 kyr. Discussions concerning the diffusion of the gases, such as  $^3\text{He}$  in quartz and the non-cosmogenic compo-

nents of the elements, addressed the present state of understanding and made suggestions for future noble gas experiments. Although M. Kurz and T. Cerling were unable to attend, references to their work in noble gas production rates are included in the bibliography.

Estimates of production rates of  $^{36}\text{Cl}$  from various pathways (thermal neutron capture, muonic reactions and spallation reactions) were presented by F. Phillips *et al.*, Swanson *et al.* and Stone *et al.* The production rate estimates varied considerably, partly due to the factors mentioned above and partly due to the complications involved in the interactions of multiple cosmogenic and non-cosmogenic production pathways. Nevertheless, the production rates of  $^{36}\text{Cl}$  are now sufficiently well known to yield ages that are within reasonably close agreement to ages calculated with cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$ , based on data presented by Ivy-Ochs *et al.*, Gosse and Klein and Phillips, F. *et al.* This agreement is encouraging for our knowledge of the relative production rates of each nuclide.

Results of ongoing numerical simulations to calculate the production of various nuclides from different targets and at different energies was presented by O'Brien, Reedy and Masarik and Reedy. Production rates from the simulations show very good agreement with some measured production rates. Uncertainties in the fluxes of cosmic-ray particles (especially neutrons and muons), uncertainties in the reaction cross sections with various target elements and other sources of error were recognized when the simulation data were evaluated. Imamura reported on measurements of cross sections for producing these nuclides by energetic neutrons. Heisinger *et al.* presented work using accelerator-produced stopped negative muons and fast muons and comparisons with a measured depth profile to 250 m. A discrepancy between the contribution of muons that some models predict and the contribution that has been previously assumed (the assumed contribution may be too high) provided insight into the importance of the muonic reactions.

### **Other Lessons Learned**

Much discussion centered on the effects of solar and Earth magnetic influences on the incident cosmic ray flux. There is a well-understood relationship between incident cosmic ray flux and the dipole moment of the Earth. Instantaneous *in-situ* cosmogenic nuclide production rates in rocks on Earth vary in a way similar to how production rates of the  $^{14}\text{C}$  in the atmosphere varies: during periods of low field intensity, production rate is high and vice versa. The main difference is that the concentration of *in-situ* nuclides is the integral of the production rates over the entire exposure period whereas  $^{14}\text{C}$  ratios record the production rate at small instances of time. Although short-lived high-amplitude changes in paleointensity will affect the atmospheric  $^{14}\text{C}$  time scale, the cosmogenic nuclide time scale will not feel the effects of a short paleointensity change unless the exposure time is of similar duration.

Several programs are being generated (Clapp *et al.*: COSMO-CALIBRATE; Dunne *et al.*) to correct cosmogenic nuclide production rates due to changes in the intensity of Earth's dipole field strength. These programs are useful to show the sensitivity of the integrated production rates of cosmogenic nuclides to changes in the geomagnetic field—however, they are only as good as the input data (see below). Using the existing paleointensity records, the programs predict that, depending on the latitude and elevation of the site, the time-integrated production rate for an exposure over the last 40 kyr is much higher (possibly ~20%) than the production rate over the last 5 kyr.

Unfortunately, existing absolute and relative paleointensity records are not sufficiently reliable to base adjustments of cosmogenic nuclide production rates. A presentation by K. Verosub and the ensuing discussions revealed the difficulties that paleomagnetists have had in interpreting the paleomagnetic data. Sources of uncertainties in the paleointensity records include poor age control, dif-



ficuity in evaluating the significance of non-dipole contributions and errors due to variations in magnetic grain size, multi-domain remanences and inclination shallowing. The uncertainty of many of the existing paleointensity records therefore probably approaches or exceeds 20%. Until more paleointensity records become available it may be difficult to adjust cosmogenic nuclide production rates with high degrees of uncertainty.

Other sources of variation in production rates were discussed at the workshop. R. Sternberg showed that significant changes in the average geomagnetic latitude of sampled sites (up to 4° over exposure durations longer than 2 kyr) are caused by secular variation of the dipole axis position. Only sites <~15 kyr are affected because the dipole axis position is approximately geocentric for longer exposures. The effects of a 2–4° uncertainty in geomagnetic latitude were modeled by Klein and Gosse, who showed that for high altitude mid-latitude sites (~30° latitude) the uncertainty could cause a discrepancy in production rate on the order of 10%.

The influence of solar modulation over a solar cycle was discussed by K. O'Brien. The model results he presented and results from simulation efforts by R. Reedy and J. Masarik suggest that solar modulation may play a significant role at high latitudes.

The uncertainty in the protocol for scaling production rates to different latitudes and altitudes was the theme of many discussions throughout the workshop. Existing data seem to suggest that the altitudinal correction of Lal (1991) is reliable to within 10%. However, J. Stone pointed out that elevation corrections may be complicated because atmospheric thickness above any site may change significantly due to a combination of factors such as eustatic sea level changes and glacial isostasy. Also, some special cases exist such as the effects on atmospheric thickness of the quasi-stable low atmospheric pressure and cold air over Antarctica. There was considerable disagreement on the reliability of the Lal (1991) latitudinal scaling. The debate over the relative significance of muon effects on latitude and altitude adjustments is not fully resolved. The effects of non-dipole field variations are presently ignored, mostly because insufficient data are available to model Quaternary non-dipole field paleointensities over the globe, according to J. Stamatakos. However, the spatial and temporal variations in non-dipole field strengths may be significant for Holocene measurements. I. Graham reported on plans that his group is making to measure production rates over a wide range of latitudes.

Discussions on the reliability of the dating methods against which the *in-situ* nuclide time scales are being calibrated centered on the <sup>14</sup>C method, and to a lesser extent, the <sup>40</sup>Ar/<sup>39</sup>Ar technique. The utility of surfaces assumed to be in saturation for <sup>10</sup>Be and <sup>26</sup>Al in Antarctica (and the reliability of the <sup>10</sup>Be and <sup>26</sup>Al half-lives) was also discussed. The majority of the production rate calibrations have been at <20 kyr, based on <sup>14</sup>C ages. The calibrated <sup>14</sup>C time for ages <~11 kyr is generally believed to be sufficiently reliable for calibration. Disagreement over the reliability of the U/Th coral calibration of older <sup>14</sup>C ages (to ~18 kyr) was not completely resolved, although M. Stuiver presented convincing isotopic data from the GISP core which supported the extended <sup>14</sup>C calibration curve. It is clear that continued improvement in calibration of the <sup>14</sup>C time scale is needed. Refinements in long term *in-situ* production rates will continue as improvements in the understanding of latitude and altitude effects enable reliable extrapolation of Antarctic production rates to other locations and by cautiously taking advantage of recent advances in Ar-Ar dating of whole rocks.

### **Suggestions for Future Calibration of Cosmogenic Nuclide Time Scales**

The following points reached general consensus among the group:

1. If possible, calibration against ages derived from other calibrated dating methods or other cosmogenic nuclide ages should be avoided. It was suggested that more emphasis should be placed on calibrating with ages based on the U and Ar systems.
2. It was pointed out that several existing production rates may be slightly low because the age assumed for the exposure duration was too high due to shielding by till or sediment (*e.g.*, rocks at Meteor Crater may have been covered by ejecta). When selecting a site for calibration, the potential for partial or complete shielding due to burial must be considered. For young sites (<20 kyr) the effects of erosion are probably insignificant when compared to the effects of burial. For example, selection of sites with glacial polish to ensure no erosion has occurred since deglaciation may be counter-productive if the reason the polish has survived is because it was protected by a soil cover.
3. The group recommended that because suitable calibration sites are so difficult to find, no specific site latitude, elevation, or exposure duration should be particularly targeted.
4. It was also agreed that future proposed calibration sites should be peer evaluated so that any merits and problems of the site can be identified before the production rate data is generated. No official forum was suggested because it was felt that informal discussion should be sufficient.
5. An attempt should be made to measure multiple nuclides at calibration sites where funding and mineralogy/lithology permit. Also, multiple samples should be analyzed for at least one nuclide at each site to help minimize the effects of erosion, burial and pre-exposure.

Participants of the workshop have also agreed to conduct an inter-laboratory comparison of various aspects of the chemistry and isotopic measurements involved in the cosmogenic nuclide methods. Wide interest was expressed in an inter-laboratory standard, with one suggestion being Tabernacle Hill, Utah, where multiple lithologies co-exist with tight age constraints.

### **Suggestions for Future Authors or Reviewers of Publications Containing Interpretations of Cosmogenic Nuclides**

In order to calculate exposure ages or erosion rate information from the measured cosmogenic isotopic abundances, numerous other measurements and data must be considered and incorporated. The protocol is generally straightforward but can be rather involved. Representatives of several of the laboratories where measurements of the abundances of the cosmogenic nuclides are made have pointed out that cosmogenic nuclide dating is beginning to be carried out by scientists who have not been involved in the technique development over the last decade. We can only recommend that the researcher considers the question being addressed before selecting the nuclide systems being utilized and that before collecting the sample, the researcher makes himself aware of all of the caveats and information needed. Once an erroneous age makes it into the literature it is sometimes very difficult to remove it.

There are several reasons why a single production rate for a particular nuclide cannot be suggested. The production rate for any nuclide at any time and location is not well known, mostly because of the very limited data set that exists but also because of small but significant discrepancies in the production rates that have been measured. Additionally, because the production rates probably change due to temporal fluctuations in the effective cosmic ray flux to the rock surface (due to past variations in geomagnetic field strength, dipole axis position, solar modulation, atmospheric pressure and

other factors), a single time-independent production rate is not viable. Finally, there remain uncertainties in the scaling of production rates over different altitudes and latitudes.

Other factors that must be considered when calculating a site production rate based on the effective cosmic ray flux a rock receives includes the rock sample thickness and depth below the surface, surface geometry (addressed by A. Gillespie), amount of shielding of cosmic radiation (*e.g.*, by snow, water, sediment and topography with respect to the horizon) and erosion rate and style.

It is therefore crucial that authors consider including the following information so that as the science continues to develop we can re-evaluate the interpretations of the cosmogenic nuclide data (*e.g.*, just as the calibration of the  $^{14}\text{C}$  timescale is continually being improved). If page length restrictions limit the ability of the incorporation of these data, then the data should be made accessible elsewhere and that source be furnished. The list is not exhaustive but we hope reviewers of manuscripts will adhere to the suggestions (Table 1). Some other data may be necessary, depending on the technique and detailed descriptions of geometry and shielding should be included if applicable.

Meaningful estimates of the uncertainties in all calculated ages and erosion rates should be provided. Because the level of confidence or types of uncertainties used to describe the error limits may vary due to nature of the data or the style of the author, we recommend that some indication of what the error limit calculation involves also be included.

TABLE 1. Information Suggested to Accompany Published Data

Data/parameter	Suggested unit or means
Site latitude <i>and</i> longitude	°
Site elevation	m
Effective attenuation length	$\text{g} \cdot \text{cm}^{-2}$
Density of the rock sampled	$\text{g} \cdot \text{cm}^{-3}$
Production rate(s) used	$\text{Atom} \cdot \text{yr}^{-1} \cdot \text{g}^{-1}$ of material
Thickness/depth of the sample	cm
Chemical data	
Isotopic data	$\text{Atom} \cdot \text{g}^{-1}$ of material, or equivalent
Geometry/slope correction applied*, †	Dimensionless
Shielding correction applied*, †	Dimensionless
Density of the covering material*	$\text{g} \cdot \text{cm}^{-3}$
Thickness of the shielding cover*, †	cm
Duration of cover if known*	yr or kyr
Erosion rate correction*, †	Dimensionless, or $\text{mm} \cdot \text{kyr}^{-1}$ or equivalent
Latitude and altitude scaling factor*, †	Dimensionless; provide ref. ( <i>e.g.</i> , Lal 1991)

\*If applicable

†Requires explanation

### Utility of Cosmogenic Nuclide Exposure History Techniques

This workshop focused on how to improve our understanding of cosmogenic nuclide production rates and make recommendations for future calibrations. However, many unresolved questions involving Neogene chronologies, landform stability and tectonic, seismic, volcanic and climatic issues can be addressed with sufficient accuracy with the present uncertainty in the rates of production of cosmogenic nuclides. Some examples include 1) estimates of erosion rates to within 20% is generally of sufficient accuracy to address landform- or basin-scale questions; 2) relative sequences of events such as detailed deglaciation patterns or fan deposition can be attained from cosmogenic

nuclides because the precision of the techniques approaches 3 to 5%; 3) similarly, cosmogenic nuclides can be used for correlation purposes (*e.g.*, the existence of Younger Dryas moraines around the world) by comparing normalized concentrations ( $\pm 10\%$ ) of the same nuclide, thereby avoiding calibration to a different time scale; and 4) chronologic questions that do not require highly accurate exposure age assignments (better than 15%), such as 1) was this colluvial deposit formed during the early Quaternary, middle Quaternary or Late Quaternary, or 2) does this moraine correspond to a marine oxygen isotope stage II or an older ice volume advance?

Although many questions can be addressed with the existing uncertainties in production rates, it is clear that better estimates of production rates are direly needed to utilize the cosmogenic nuclide techniques to their full potential. A number of challenging questions that directly impact society will remain largely unanswered until the cosmogenic nuclide methods evolve further over the next decade.

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## SANTA FE WORKSHOP ABSTRACTS<sup>1</sup>

### REFINING ESTIMATES OF <sup>10</sup>Be AND <sup>26</sup>Al PRODUCTION RATES

PAUL BIERMAN,<sup>2</sup> PATRICK LARSEN,<sup>2</sup> ERIK CLAPP<sup>3</sup> and DOUGLAS CLARK<sup>4</sup>

We have taken a variety of approaches to refine the existing <sup>10</sup>Be and <sup>26</sup>Al production rate estimates of Nishiizumi *et al.* (1989), including measuring nuclide abundances on boulders and bedrock surfaces exposed on the relatively well-dated Laurentide moraine, making altitude and latitude corrections using different, now currently accepted protocols, and modeling the effect of changing geomagnetic field strength on production rates over time.

In north-central New Jersey, we collected 16 samples of quartzite and gneiss from striated and/or glacially molded outcrops and large glacial erratics on or just behind the terminal moraine of the Laurentide ice sheet (Larsen 1995). The exposure age of the moraine is estimated to be ~21.5 ky sidereal years on the basis of several limiting <sup>14</sup>C ages. The calibration of these <sup>14</sup>C ages is based on the recommendations of Stuiver *et al.* (1991). Average concentrations and standard errors of the mean of  $1.37 \pm 0.04 \times 10^5$  (n=12) atoms g<sup>-1</sup> SiO<sub>2</sub> for <sup>10</sup>Be and  $8.03 \pm 0.29 \times 10^5$  (n=14) atoms g<sup>-1</sup> SiO<sub>2</sub> for <sup>26</sup>Al can be used to calculate integrated sea level, high-latitude production rates and associated standard errors of the mean of  $5.17 \pm 0.15$  and  $30.40 \pm 1.01$  atoms g<sup>-1</sup> yr<sup>-1</sup> for <sup>10</sup>Be and <sup>26</sup>Al, respectively. The isotope abundances show no significant variation between boulder and outcrop sample locations and quartzite and gneiss sample lithologies, implying that the effects of erosion and till cover are negligible. We have scaled these abundance data to sea level and high latitude using the neutron data of Lal (1991) following the finding by Brown *et al.* (1995) that muon production is insignificant; however, because our samples were collected at relatively high latitude (41°) and low altitude (300 m), the choice of scaling protocol is unimportant. The integrated production rates calculated over the past 21.5 ky are *ca.* 18% lower than previously determined integrated production rates over the 11 ky (Nishiizumi *et al.* 1989) and 50 ky (Nishiizumi *et al.* 1991b) time frames.

Comparing these three different production rate estimates is not straightforward. Each was calculated from samples exposed for varying lengths of time at different altitudes and latitudes. Scaling the lower-latitude, higher-altitude measurements from the Sierra Nevada and Meteor Crater to sea level and high latitude involves choosing an effective geomagnetic latitude for the duration of exposure, determining the contribution of muons to <sup>10</sup>Be and <sup>26</sup>Al production as a function of altitude and latitude, and determining how instantaneous production rates have changed over the duration of exposure at each site.

We find that by assuming muon production is inconsequential at sea level (Brown *et al.* 1995), scaling the three data sets for spallation only, and incorporating a geomagnetically driven production rate forcing model (Clark *et al.* 1995; Clapp and Bierman, in review), the three different production rate estimates can be reconciled to within several percent if the exposure age of the Sierra Nevada calibration sites is taken to be 13.5 ky as suggested by Clark *et al.* (1995). Although such agreement is promising, additional data must be gathered to test these findings. Until those data have been gathered, nuclide production rates as a function of altitude, latitude and exposure duration will remain a significant uncertainty in the interpretation of nuclide abundance data.

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## AN ANTARCTIC PERSPECTIVE ON *IN-SITU* COSMOGENIC NUCLIDE PRODUCTION

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Long-term average production rates of some nuclides can be constrained by examining slowly eroding, old rock surfaces. At steady state with respect to production and radioactive decay, production rates are simply calculated from concentration data (*e.g.*,  $P=N\lambda$ ), as long as erosion is negligible (Brook *et al.* 1995). The possibility that erosion is non-negligible makes these production rates minimum values. Examination of our and other published <sup>26</sup>Al data (half-life  $7.2 \times 10^5$  yr) from a total of 15 Antarctic rock surfaces with <sup>10</sup>Be exposure ages older than 2 Ma (and therefore model erosion rates  $< \sim 7$  cm/Ma) yields <sup>26</sup>Al production rates that agree well with the published Sierra Nevada rates (Nishiizumi *et al.* 1989), assuming the altitude/latitude scaling of Lal (1991). The samples span an altitude range of 1380 to 2650 m and at each sample altitude the calculated production rates agree with the scaled Sierra Nevada rates within  $\sim 10\%$ . The sea level production rate derived from the data (Lal 1991) is  $35 \pm 2$  at/g/yr, close to the predicted value (Lal 1991) of 37 at/g/yr. These observations suggest that the long-term average <sup>26</sup>Al production rate is not higher than, and is probably close to, the value determined for glaciated bedrock in the Sierra Nevada exposed over the last  $\sim 11$  ka<sup>2</sup>. The results also imply that the scaling factors (Lal 1991) are accurate within the latitude and altitude range considered here (excepting the possibility of compensating errors). As there is no reason to expect temporal variations in the <sup>26</sup>Al/<sup>10</sup>Be production ratio, the long-term <sup>10</sup>Be production rate is also probably close to the Sierra Nevada rate (Nishiizumi *et al.* 1989). These conclusions are similar to those of Nishiizumi (this issue).

We have also collected two 1–1.5 m drill cores in Antarctic sandstone bedrock to examine the depth dependence of <sup>10</sup>Be, <sup>26</sup>Al, and <sup>3</sup>He production. <sup>10</sup>Be and <sup>26</sup>Al profiles from one core were reported previously (Brown *et al.* 1992). Both cores have exponential profiles; scale lengths are close to expected values and are  $152 \pm 5$  and  $145 \pm 5$  g/cm<sup>2</sup> for <sup>10</sup>Be and  $153 \pm 13$  and  $152 \pm 5$  g/cm<sup>2</sup> for <sup>26</sup>Al. Contrary to the calculations of Nishiizumi *et al.* (1989), these data indicate that muons produce  $< 1\text{--}3\%$  of total <sup>10</sup>Be and <sup>26</sup>Al at the altitudes and latitude of these cores, consistent with previous conclusions (Brown *et al.* 1995) based on a depth profile of <sup>10</sup>Be at low altitude near the equator.

The situation for <sup>3</sup>He is more complicated. One of the two cores exhibits an exponential decrease in cosmogenic <sup>3</sup>He with a scale length of  $\sim 150$  g/cm<sup>2</sup>. The second has a distinctly higher scale length,  $227 \pm 14$  g/cm<sup>2</sup>, over a similar depth interval. Studies of different size quartz grains in each core show that the discrepancy, which can be thought of as “extra <sup>3</sup>He” at depth, or loss of <sup>3</sup>He at the surface, is not an artifact of diffusion. It also does not appear to be caused by the presence of a non-cosmogenic <sup>3</sup>He component. Production of <sup>3</sup>He by muons is a remaining possibility. A model that includes the processes of erosion, diffusion, and <sup>3</sup>He production by neutrons and muons can approximately reproduce the observed profile with reasonable parameters, if exposure times are very long (*e.g.*, of order 20 Ma or greater), and if production of muons is  $\sim 10\%$  of total production at 1700 m. While we are uncertain if this explanation of our data is correct, our observations suggest that further investigation of production rates of <sup>3</sup>He due to muon interactions is warranted.

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## DIRECT EXAMINATION OF TERRESTRIAL COSMOGENIC NUCLIDE PRODUCTION RATES OF $^{10}\text{Be}$ , $^3\text{He}$ AND $^3\text{H}$

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We have determined production rates of  $^{10}\text{Be}$ ,  $^3\text{He}$ , and  $^3\text{H}$  in tanks of water exposed for a period of 15 months at a range of altitudes (620 m, 3810 m, and 4745 m) in the French Alps. Measurements of  $^3\text{He}$  and  $^3\text{H}$  were undertaken by static mass spectrometry at Laboratoire de Modelisation du Climat et de l'Environnement in Saclay, France, while  $^{10}\text{Be}$  determinations were made at the Tandatron Accelerator Mass Spectrometry Facility in Gif-sur-Yvette, France. Corrections were made for low levels of  $^{10}\text{Be}$  and  $^3\text{H}$  present in the water, for incomplete degassing of atmospheric He, and for production of the three nuclides within the tanks in the laboratory before and after installation at altitude. Further minor corrections were made for self-shielding within the tanks and for skyline shielding by local topography. Our results indicate production ratios for oxygen spallation of  $\sim 2:1$  for  $^3\text{He}:^3\text{H}$  and  $\sim 20:1$  for  $(^3\text{He}+^3\text{H}):^{10}\text{Be}$ . Strategies for application of our production rates to longer time periods and other locations, as well as to specific mineral phases, will be discussed.

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## COSMO-CALIBRATE: A PROGRAM FOR CALIBRATING COSMOGENIC EXPOSURE AGES

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In order to account for geomagnetically induced changes in cosmogenic nuclide production rates *in situ*, and thereby increase the potential accuracy of exposure age dating, we have developed a numerical model that generates geomagnetically calibrated exposure ages. Currently accepted, model-age calculations are based on time-averaged production rates. Use of time-averaged production rates may introduce systematic errors >20% in exposure ages causing cosmogenically dated events to be correlated incorrectly with events dated by other means (Clark *et al.* 1995). Applying our model to existing nuclide abundance data generally increases calculated exposure ages (Fig. 1), supports the  $^{10}\text{Be}$ -based assertion of Gosse *et al.* (1995) that a glacial advance in the Rocky Mountains may have occurred during the Younger Dryas, and most importantly, reconciles three apparently disparate production rate estimates for  $^{26}\text{Al}$  and  $^{10}\text{Be}$ .

We have created a computer program (COSMO-CALIBRATE) that uses generally accepted geomagnetic paleointensity records (Meynadier *et al.* 1992; McElhinny and Senanayake 1982) and empirical relationships to account for cosmogenic isotope production rate variations over the last 140 kyr. Magnetic field strengths are converted to apparent paleolatitudes using the formulation of Nishiizumi *et al.* (1989) after which instantaneous production rates are calculated using the third degree polynomial of Lal (1991). Muon contribution is currently assumed to be minimal, as suggested by Brown *et al.* (1995). Samples exposed at lower latitudes and high altitudes will have experienced greater production rate variation (Fig. 2).

Our program calculates instantaneous  $^{10}\text{Be}$  and  $^{26}\text{Al}$  production rates at any given sample site (altitude and latitude) and outputs both geomagnetically calibrated and uncalibrated exposure ages from sample isotopic abundance data. Calibration, such as we propose, will likely increase the accuracy of exposure ages and once verified by additional data, may allow for more robust cosmogenic dating and correlation of relatively brief geomorphic and climatic events. Our program is available as user-friendly, compiled Macintosh code by anonymous ftp from `beluga.uvm.edu`.

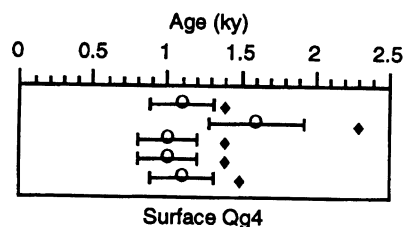


Fig. 1. Calibrated  $^{10}\text{Be}$  exposure ages from COSMO-CALIBRATE 1.7 (diamonds) and uncalibrated  $^{10}\text{Be}$  exposure ages (Bierman *et al.*) calculated using production rates of Nishiizumi *et al.* (1989), for 11,000 cal yr BP (open circles).

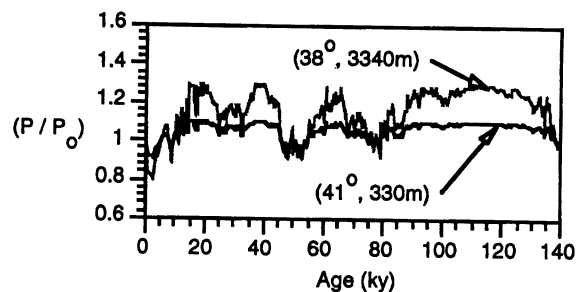


Fig. 2. Relative instantaneous production rates calculated by COSMO-CALIBRATE for two study sites: Sierra Nevada ( $38^\circ$ , 3340 M) and the Laurentide Terminal Moraine ( $41^\circ$ , 330 m)

## $^{10}\text{Be}$ AND $^{26}\text{Al}$ PRODUCTION RATES AND A REVISED GLACIAL CHRONOLOGY FOR THE SIERRA NEVADA

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New  $^{14}\text{C}$  ages for Sierra Nevada deglaciation suggest that currently accepted late-Pleistocene production rates of *in-situ* cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$  are  $\sim 20\%$  too high because the assumed age of exposure is too young. The original production rates for the isotopes were calibrated based on measurements of glacially polished granite from the Sierra Nevada, assumed to have been deglaciated  $\sim 11,000$  calibrated (or sidereal) years ago (11,000 cal yr BP) (Nishiizumi *et al.* 1989). This exposure age was estimated from minimum-limiting conventional radiocarbon ages of  $\sim 10,000$   $^{14}\text{C}$  yr BP for basal bulk sediments from two sites, a lake and a meadow, dammed behind Tioga (last late-Wisconsin maximum) recessional moraines (Adam 1967; Mezger and Burbank 1986).

Calibrated ages (Bard *et al.* 1990; Stuiver and Reimer 1993) of 12 new  $^{14}\text{C}$  dates, of basal or near-basal lake sediments from cores of ten postglacial lakes, show instead that Sierra Nevada deglaciation was underway by 16,000–19,000 cal yr BP ( $\sim 13,500$ – $16,000$   $^{14}\text{C}$  yr BP) and that the range was effectively deglaciated before  $\sim 13,100$  cal yr BP (11,190  $^{14}\text{C}$  yr BP) (Clark *et al.* 1995; Clark and Gillespie, in press). The coring sites all lie inside the maximum Tioga ice limits, as do the two cited by Nishiizumi *et al.* (1989), and thus provide minimum ages for onset of deglaciation. The highest sites near the crest of the range, however, provide minimum ages for complete retreat of Tioga glaciers and thus minima for the exposure age of sites sampled by Nishiizumi *et al.* for calibration. The highest lakes we cored, in the headwaters of Bishop Creek, are formed behind moraines of the Recess Peak advance, which ended before 13,100 cal yr BP (Clark and Gillespie, in press). We have

mapped and correlated Recess Peak moraines along the crest of the Sierra from southern to northern limits of the advance, and find that all calibration sites of Nishiizumi *et al.* lie downstream from Recess Peak moraines in those drainages. Thus, our minimum age control for the Recess Peak advance in Bishop Creek also provides a firm minimum age for exposure of the calibration sites. The accuracy of the AMS  $^{14}\text{C}$  dates from the Bishop Creek cores is supported by: 1) other dates from higher in the same cores that are internally consistent; 2) parallel dates between adjacent gyttja and macrofossils in the cores that are indistinguishable within 1-sigma analytic error (typically <1%); 3) similar findings in dates of cores from other areas that also are internally consistent; and 4) absence of sources of contamination, especially "old" carbon, in the drainage basins upstream from the lakes. These observations substantiate the stability of carbon in high Sierran lake basins. Together, the new limiting  $^{14}\text{C}$  dates indicate that Tioga deglaciation occurred at least 2000 cal yr before 11,000 cal yr BP.

Recalculating the production rates using the formulation and measured abundances of Nishiizumi *et al.* (1989) but assuming the sites were deglaciated 13,500 cal yr BP yields sea-level, high-latitude production rates that are ~20% lower than in the original calibration for both isotopes. This difference agrees within several percent of an independent late-Pleistocene calibration from the Laurentide terminal moraine in New Jersey (Larsen 1995), if appropriate latitudinal scaling and the affects of geomagnetic field-strength fluctuations on production rates are considered (Clark *et al.* 1995; Clapp and Bierman 1995). Our results emphasize the need for accurate, independent age control and geologic context as primary foundations for production-rate calibrations of cosmogenic nuclides.

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## PLANS FOR CALIBRATION OF THE COSMIC RAY FLUX OVER THE PAST 500,000 YEARS USING VOLCANIC ROCK

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While recent measurements of neutron fluxes can be combined with nuclear reaction data using Monte Carlo simulation techniques to predict the current production rates of cosmogenic nuclides, such techniques cannot be used to explore the past history of the cosmic ray flux (CRF). Although certain effects are fairly well understood or modeled, such as short-term cycling of the CRF at the surface of the Earth in response to variations in the Earth's magnetic field, other variations are not as well categorized. While some ice cores and similar historical record-keepers provide some insight into the past, it is the intent of this project to calibrate the method of  $^{36}\text{Cl}$  rock exposure dating, and hence the cosmogenic production rates, back through 500 ka by comparing the  $^{36}\text{Cl}$  concentrations with  $^{40}\text{Ar}/^{39}\text{Ar}$  data in collected samples from the central Andes.

The central Andes is an ideal location for a study such as this. This hyper-arid and isolated environment offers samples that have remained undisturbed and effectively uneroded in a location that has had little or no water or vegetation cover. In addition, the high altitude of the region insures very high nuclide concentrations. The effectively instantaneous creation of the lava flows aids in correlating the  $^{40}\text{Ar}/^{39}\text{Ar}$  to the measured  $^{36}\text{Cl}$  concentrations, and the long-term development of the volcano by successive flows will provide a moderately wide span of dates for the calibration. As an additional incentive, Tata Sabaya, the specific volcano to be studied, underwent a cone collapse that resulted in an easily identifiable debris flow. This extra "instantaneous" event provides another point on the calibration curve.

When combined with the existing calibration data, which focus on more recent time periods, these new data should provide a broader perspective on long-term variations in the CRF. Analysis will incorporate Monte Carlo neutron transport simulations to account for rock geometry and water content in addition to the standard scaling factors such as geometric shielding, altitude and latitude variations, and erosion effects. With the higher frequency variations from magnetic field fluctuations effectively averaged out after only a few cycles, any long-term variations in the CRF, and hence, the radioisotope production rates, will be identifiable, making the results of the project useful to any method or procedure that relies on cosmogenic nuclides.

#### **PRODUCTION RATE OF *IN-SITU* COSMOGENIC $^{10}\text{Be}$ IN QUARTZ AT HIGH ALTITUDE AND MID LATITUDE**

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*and*

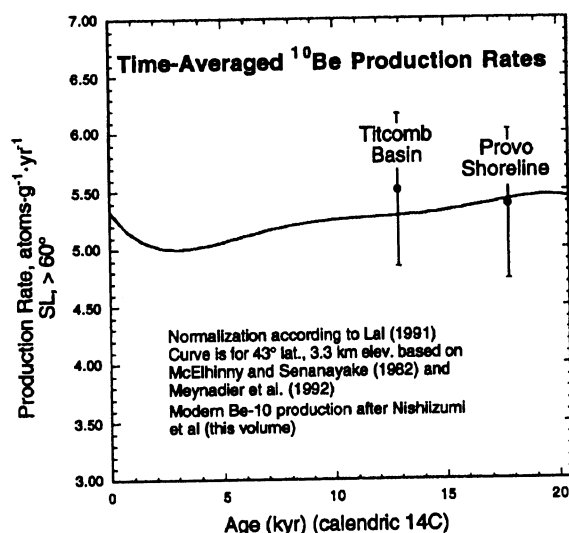
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Several considerations must be made when attempting to empirically calibrate a cosmogenic nuclide time scale to the calendric time scale. The sampled surface should not have been buried or eroded during exposure, the initial cosmogenic concentration of the nuclide must be zero (*i.e.*, no inheritance from prior exposure), there should be no significant shielding or geometry problems, and the lithology must be suitable. The duration of the exposure of the surface must also be accurately known. Finally, the geomagnetic latitude and duration at which calibrations are done should be taken into account because at some latitudes even a small uncertainty in geomagnetic latitude can produce a large uncertainty in the production rate. For calibration sites having <10 kyr exposure, the uncertainty in geomagnetic latitude due to pole position secular variation may be a factor because the dipole axis may not have been geocentric. If the change in production rate due mainly to geomagnetic field intensity variation is the focus, it may be useful to concentrate our measurement of production rates at sites within the latitudinal zone ( $\sim 15^\circ$ – $45^\circ$ ) where these geomagnetic uncertainties have the most significant effects.

We have sampled 10 boulders from the inner Titcomb Basin moraine in the Wind River Range, Wyoming (3.3 km,  $43^\circ\text{N}$ ), which has been correlated to the Temple Lakes moraine, dated by Zielinski and Davis (1987) and Davis (1994), at  $\sim 11.4$  to  $11.7$   $^{14}\text{C}$  kyr. The site production rate of  $^{10}\text{Be}$  in quartz assuming 12.9 cal kyr (Stuiver and Reimer 1993) for the moraine age is  $51.7$  atoms  $\cdot$   $\text{g}^{-1}$   $\text{yr}^{-1}$ ,  $53.6$  atoms  $\cdot$   $\text{g}^{-1}$   $\text{yr}^{-1}$  if adjustments are made for the effects of snow and one of the ten samples is considered an outlier. The production rate at sea level, high latitude (using Lal 1991) is *ca.*  $5.5$  atoms  $\cdot$   $\text{g}^{-1}$

$\text{yr}^{-1}$  based on nine samples and adjusted for snow cover. Considering the uncertainties in the exposure time, snow density, and muonic scaling, the uncertainty in this production rate is  $\pm 12\%$ .



Preliminary results based on  $^{10}\text{Be}$  measurements of two samples collected from a Provo Lake wave cut shoreline (1.5 km, 41°N) will also be presented. The results suggest the time-averaged  $^{10}\text{Be}$  production rate for 14.4  $^{14}\text{C}$  kyr (Oviatt, Currey and Sak 1992; 17.6 cal kyr) of exposure at sea level >60° latitude is  $5.4 \pm 0.6$  atoms · g<sup>-1</sup> yr<sup>-1</sup> (uncertainty assigned as above).

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#### THE $^{21}\text{Ne}$ PRODUCTION RATE IN A SI TARGET AT MOUNTAIN ALTITUDES

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The isotopic signature of cosmic ray produced Ne was first measured in Antarctic rocks (Graf *et al.* 1991; Niedermann *et al.* 1993). Production rates of cosmogenic Ne were determined in quartz from glacially polished Sierra Nevada rocks which were exposed for only *ca.* 10<sup>4</sup> yr (Niedermann *et al.* 1994). However, past and present production rates of  $^{21}\text{Ne}$  and other relevant cosmic ray produced nuclides are still poorly known. Temporal variations in the production rates can be studied by determinations of the products in well dated rocks and minerals thereof. The present production rates can



be measured on exposed targets at mountain altitudes. We report first  $^{21}\text{Ne}$  results obtained from a Si target exposed on Mt. Evans, Colorado.

We used an experimental melt (Corning Glass) substituting a small amount of Li for Na. The glass (2091 g) was placed in a  $13 \times 31$  cm stainless steel cylinder. The stem was double valved to prevent leakage. To reduce self-shielding, the cylinder was designed to be only about half filled and lie on its side. The glass was degassed by heating numerous times above the softening temperature. Reaction of the glass with the stainless steel cylinder was apparent by glass analysis after the Ne study. The sample was exposed to cosmic rays for *ca.* 3 yr on the summit of Mt. Evans, Colorado before it was transported to La Jolla for analysis.

Calibrations of the  $^{40}\text{Ar}^{++}$  and  $\text{CO}_2^{++}$  interferences on masses 20 and 22, respectively, revealed that the ratios of doubly to singly charged  $^{40}\text{Ar}$  and  $\text{CO}_2$  ions are not constant. Our Ne analytical procedure monitors the peaks at masses 2, 18, 19, 20, 21, 22, 40, 42, and 44. The ratios  $^{40}\text{Ar}^{++}/^{40}\text{Ar}^+$  and  $\text{CO}_2^{++}/\text{CO}_2^+$ , respectively, are calculated according to Graf *et al.* (1994) and the appropriate calibrations. The resulting interference corrections are subtracted individually for each cycle. We also need to calibrate the Ne sensitivity of the instrument as changes in the charge states and charge density in the ion source affect concentration measurements.

A significant fraction of the cosmic-ray produced Ne (36%) was in the gas phase of the cylinder. Ne was then extracted in 3 steps at increasing temperatures and a re-extraction step at  $950^\circ\text{C}$  was added. Trapped Ne in all steps was fractionated up to 2% per amu, an effect that is presumably due to the extensive degassing prior to exposure. The total excess  $^{21}\text{Ne}$  observed in the glass corresponds to 1500 atoms  $\text{g}^{-1}$  ( $\text{SiO}_2$ ) and we calculate a production rate  $P_{21}(\text{SiO}_2) = 410 \pm 60$  atoms  $\text{a}^{-1}\text{g}^{-1}$  ( $\text{SiO}_2$ ) at Mt. Evans (4250 m) altitude. The uncertainty includes current uncertainties in the Ne sensitivity and in mass discrimination which were added quadratically. The procedure of Lal (1991) is used to calculate the production at sea level, and  $P_{21}(\text{SiO}_2) = 19.3$  atoms  $\text{a}^{-1}\text{g}^{-1}$  ( $\text{SiO}_2$ ) for latitudes  $>60^\circ$  is obtained. No corrections have yet been applied for self-shielding and for flux variations during the solar cycle (the exposure occurred during maximum solar activity). This value may be compared to a production rate  $P_{21} = 21$  atoms  $\text{a}^{-1}\text{g}^{-1}$  (quartz) obtained by Niedermann *et al.* (1994) for Sierra Nevada quartz samples.

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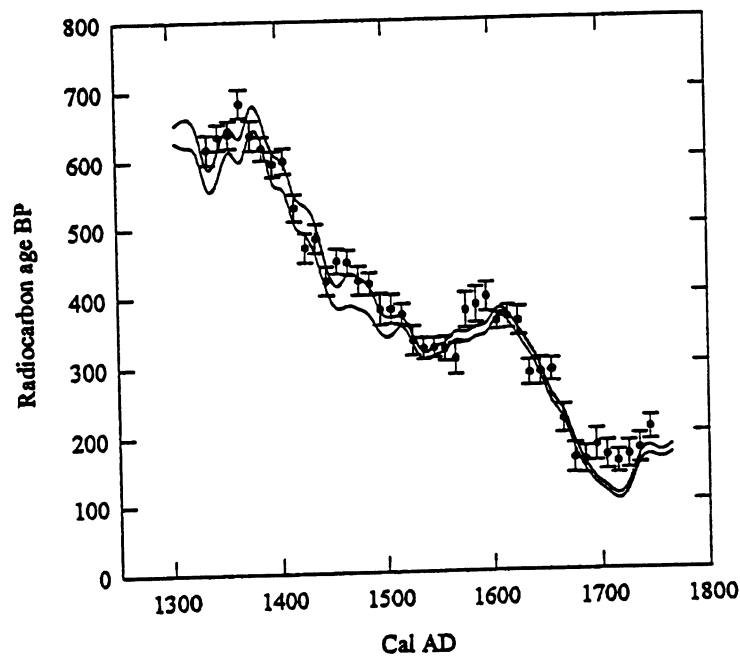
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## RECENT AND PLANNED RESEARCH IN COSMOGENIC ISOTOPES AT GNS

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The AMS laboratory at GNS was set up some ten years ago and has established an international reputation in  $^{14}\text{C}$  analysis directed mainly towards archaeometry and atmospheric studies. A recently obtained calibration curve has provided a more secure basis for radiocarbon measurement. Tree rings from a section of native *Matai* covering the period AD 1335 to 1745 have been dated and used to generate a  $^{14}\text{C}$  calibration for Southern Hemisphere wood. Comparison of this curve with calibration data for Northern Hemisphere woods shows no systematic difference.



Research into other cosmogenic isotopes ( $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$ ) at GNS is still in its infancy. Analysis of  $^{10}\text{Be}$  in marine sediments, loess and Antarctic soils has yielded some interesting but inconclusive results. Planned research for 1996 involves direct measurement of  $^{10}\text{Be}$  fluxes and *in-situ* production rates across a wide range of Southern Hemisphere geomagnetic latitudes ( $3^\circ\text{S}$  to  $79^\circ\text{S}$ ). On-site measurement of neutron and/or muon flux is also planned. Sampling sites have been investigated and some preliminary rain collections made and measured.

## DEPTH PROFILES OF COSMOGENIC NUCLIDES AND ACCELERATOR SIMULATION EXPERIMENTS WITH STOPPED NEGATIVE MUONS AND FAST MUONS

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*In-situ* production of cosmogenic nuclides is important for the determination of background events in all low-level detection experiments, e.g., cryogenic dark matter detectors or geochemical experiments such as the geochemical solar neutrino experiment  $^{205}\text{Tl}(\nu_e, e^-)^{205}\text{Pb}$  (Neumaier, Nolte and Morinaga 1991), and in many geophysical applications such as the determination of erosion rates.

The *in-situ* production of radionuclides has been calculated as the function of the depth  $z$ , taking into account spallation reactions, reactions with captured negative muons, reactions with fast muons and background reactions. The production rate due to  $\mu^-$  capture can be expressed by  $P_{\mu^-}(z) = I_{\mu^-}(z) \cdot f_C \cdot f_D \cdot f^*$  with the rate of stopped negative muons  $I_{\mu^-}(z)$ , the chemical compound factor  $f_C$ , the probability of nuclear capture  $f_D$  and the probability  $f^*$  of the reaction channel after nuclear  $\mu^-$  capture to the investigated nuclide or isomeric state.

The production due to fast muons is given by  $P_{\mu, \text{fast}}(z) = \Phi_{\mu, \text{fast}} \cdot \sigma$  with the flux of fast muons  $\Phi_{\mu, \text{fast}}$ . The energy-dependent cross section  $\sigma$  is given by  $\sigma = \sigma_0 \cdot E_{\text{mean}}^{0.7}$  according to the Wolfendale rule with the mean muon energy  $E_{\text{mean}}$  in GeV at the depth considered.

These contributions due to  $\mu^-$  capture and fast muons were studied by accelerator simulation experiments at PSI, Villigen, with slow negative muons and at CERN with 190 GeV muons. The following probabilities  $f^*$  of particle emission channels after  $\mu^-$  capture have been measured in addition to earlier measurements (Dockhorn *et al.* 1991, Strack *et al.* 1994):  $f^* = (5.2 \pm 0.6) \cdot 10^{-3}$  for  $\text{O}(\mu^-, \nu_{\mu} \alpha \text{pxn})^{10}\text{Be}$  [ $x = 1-3$ ], and  $f^* = (1.4 \pm 0.4) \cdot 10^{-3}$  for  $\text{S}(\mu^-, \nu_{\mu} \alpha \text{xn})^{26}\text{Al}$  [ $x = 2-4, 6$ ]. From the result of the oxygen irradiation and the earlier result obtained for  $^{26}\text{Al}$  from the irradiation of silicon, the production ratio of  $^{26}\text{Al}$  to  $^{10}\text{Be}$  after  $\mu^-$  capture in quartz has been deduced to be  $P(^{26}\text{Al})/P(^{10}\text{Be}) = 7.3 \pm 1.2$  (Heisinger *et al.*). For the determination of the fast muon cross section  $\sigma_0$  of  $^{10}\text{Be}$  (in O),  $^{14}\text{C}$  (in O),  $^{26}\text{Al}$  (in Si, S and Al),  $^{36}\text{Cl}$  (in Ca),  $^{53}\text{Mn}$  (in Fe) and  $^{205}\text{Pb}$  (in Tl) several targets have been irradiated at CERN. This work is still in progress.

In addition, natural depth profiles of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in quartz have been measured in Northern Bavaria up to depths of 250 m and have been compared, taking erosion into account, with calculated profiles. From this comparison, the erosion rate for the last million years has been determined to be ca.  $5 \mu\text{m yr}^{-1}$  (Heisinger *et al.*).

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## NEUTRON-INDUCED PRODUCTION CROSS SECTIONS FOR LONG-LIVED NUCLIDES AND RARE GASSES: SOME EXPERIMENTAL DATA

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We present data on neutron-induced nuclear cross sections for the production of long-lived radionuclides of  $^3\text{H}$ ,  $^{7,10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$  and rare gases  $^{21,22}\text{Ne}$ , which have been measured by our group in the last several years, using semi-monoenergetic neutrons from the  $^9\text{Be}(p,n)$  and/or  $^7\text{Li}(p,n)$  reactions.

Cosmogenic nuclide production rates depend on chemical composition of the sample material and on particle fluxes of cosmic ray secondaries. Neutrons with energies up to a few hundreds MeV are the major contributing particles in terrestrial environments. Since the neutron energy spectrum depends considerably on the chemical composition of surrounding materials, knowledge of cross sections is indispensable for understanding the production rates in various terrestrial environments. Production cross sections are determined in most cases by counting the number of atoms produced from the known number of target atoms by irradiating with a known particle fluence. Because of the absence of an appropriate monoenergetic source, there have been very few experimental data on neutron-induced cross sections for energies above 20 MeV. Above 20 MeV, the most practical way is to use semi-monoenergetic neutrons from secondary emission through the  $^9\text{Be}(p,n)$  and  $^7\text{Li}(p,n)$  reactions.

We have two facilities available for semi-monoenergetic neutron irradiation: the SF cyclotron facility at the Institute for Nuclear Study (INS), University of Tokyo for 10–38 MeV, and the AVF cyclotron facility at the Japanese Atomic Energy Research Institute (JAERI) in Takasaki for 40–90 MeV (Nakamura *et al.* 1994). In addition, we have used the Van de Graaff accelerator at the Fast Neutron Laboratory (FNL) at Tohoku University for <1–16 MeV monoenergetic neutrons. Because of the limited machine time, many of the cross section studies are still underway. In the workshop I will present the data (including the published ones) on production of  $^3\text{H}$  from oxygen (<40 MeV),  $^{7,10}\text{Be}$  from C, N and O (<70 MeV),  $^{26}\text{Al}$  from Al and Si (<40 MeV),  $^{21,22}\text{Ne}$  from Mg and Al (<40 MeV).

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## A COMPARISON OF $^{10}\text{Be}$ , $^{26}\text{Al}$ , $^{36}\text{Cl}$ IN BOULDERS FROM A YOUNGER DRYAS MORaine, JULIER PASS, GRAUBÜNDEN, SWITZERLAND

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$^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{36}\text{Cl}$  measured in six boulders from an Egesen moraine complex at Julier Pass, Switzerland, gave a mean age of *ca.* 11,000 yr. Radiocarbon dating, geomorphologic evidence and regional correlation constrain Egesen moraines to have been formed as Alpine glaciers advanced during the Younger Dryas (YD) cold event (11,000 to 10,000  $^{14}\text{C}$  yr ago). Corrections for thickness of sample and topographic shielding have been made, but snow and erosion corrections have not.

With the latitude and altitude corrections stated in Lal (1991) as a basis, production rates for the three isotopes can then be examined. Overall, the agreement of the surface exposure ages from different isotopes in single boulders, between all six boulders and with the known age of the YD is very good. This leaves little room for significant changes in production rates over the last *ca.* 12,000 yr and indicates that present production rates (Nishiizumi *et al.* 1989; Phillips *et al.* 1996) are not that far off.

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## PRODUCTION OF $^{14}\text{C}$ AND $^{10}\text{Be}$ IN THE ATMOSPHERE AND ON THE EARTH'S SURFACE

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In recent years, there has been growing interest in the production of cosmogenic nuclides in rock surfaces as well as in the atmosphere, because of possible applications to a variety of fields. Geophysical and temporal variations in the production rates of cosmogenic nuclides must be understood in order to be useful for such diverse applications as estimating the exposure ages of a geomorphic surface based on *in-situ*-produced cosmogenic nuclides and calibration of  $^{14}\text{C}$  ages.

Jull *et al.* (1988) reported the first measurements of cosmogenic  $^{14}\text{C}$  in terrestrial rocks based an extension of earlier work on meteorites (Jull *et al.* 1989). Measurable levels of *in-situ*-produced cosmogenic  $^{14}\text{C}$  have been found in rocks collected from 3300 to 5452 m from North and Central America and Hawaii (Jull *et al.* 1988, 1992, 1994; Donahue, Jull and Toolin 1990). These measurements allowed us to estimate differences in sea-level production rate on the basis of latitude. Temporal changes in atmospheric and *in-situ*  $^{14}\text{C}$  production rates are also significant. Tree-ring data suggest that substantial variations in the production rate of  $^{14}\text{C}$  in the atmosphere occur (Damon and Sonett 1991). We know that the production 10,000 yr ago was of the order of 10% higher than today. Bard *et al.* (1990) argue for larger variations of up to 40% in the Late Pleistocene, based on comparisons of  $^{14}\text{C}$  ages with U-Th ages of corals. Long-term variations in the tree-ring  $^{14}\text{C}$  are probably due to geomagnetic variations, although short-term variations are thought to be solar (Damon and Sonett 1991). Bard *et al.* (1990) argue that same geomagnetic influences can explain the larger  $^{14}\text{C}$  effects seen in late Pleistocene coral results. Edwards *et al.* (1993) studied  $^{14}\text{C}$  in Late Pleistocene corals and found larger effects than can be ascribed to geomagnetic effects. Oceanic effects must have a strong, if not dominant, effect on atmospheric  $^{14}\text{C}$  variations.

For *in-situ* produced isotopes, however, geomagnetic effects are expected to predominate. Estimating geomorphic surface exposure ages or erosion rates using *in-situ* cosmogenic nuclide measurements requires us to know the average production rate over the exposure time of the surface. The record of  $^{10}\text{Be}$  produced in the atmosphere, observed in deep-sea sediments and ice, gives us a possible way of estimating these integral variations. Integration of  $^{10}\text{Be}$  in a marine sediments suggests that the effect of "spikes" at 30, 43 and 60 Ka due to cosmic-ray fluctuations possibly due to supernova or other transient phenomena (McHargue *et al.* 1995) will not have much effect on  $^{14}\text{C}$ . The total effect on *in-situ*-produced  $^{14}\text{C}$  is <10%. Similarly, such "spikes" cannot explain the deviations of  $^{14}\text{C}$  and U-Th observed by Bard *et al.* (1990), which are probably a combination of oceanic and

geomagnetic influences. However, transient changes in the cosmic-ray flux would likely have greater effects on longer-lived nuclides.

TABLE 1. Production Rates of *In-Situ*-Produced  $^{14}\text{C}$  as a Function of Latitude

Sample	Geomagnetic latitude	P at sea level ( $^{14}\text{C}/\text{g}/\text{yr}$ )	Reference
Mt. Massive, Colorado	49°N	21 ± 2	Jull <i>et al.</i> (1988); Donahue, Jull and Toolin (1990)
Tabernacle Hill, Utah	49°N	20 ± 2	Jull <i>et al.</i> (1992)
La Malinche, Mexico	28°N	16 ± 2	Jull <i>et al.</i> (1988)
Popocatepetl, Mexico	28°N	12 ± 2	Jull <i>et al.</i> (1988)
Mauna Loa, Hawaii	18°N	15 ± 2	Donahue, Jull and Toolin (1990)

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#### TERRESTRIAL FACTORS THAT INFLUENCE PRODUCTION RATES

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and

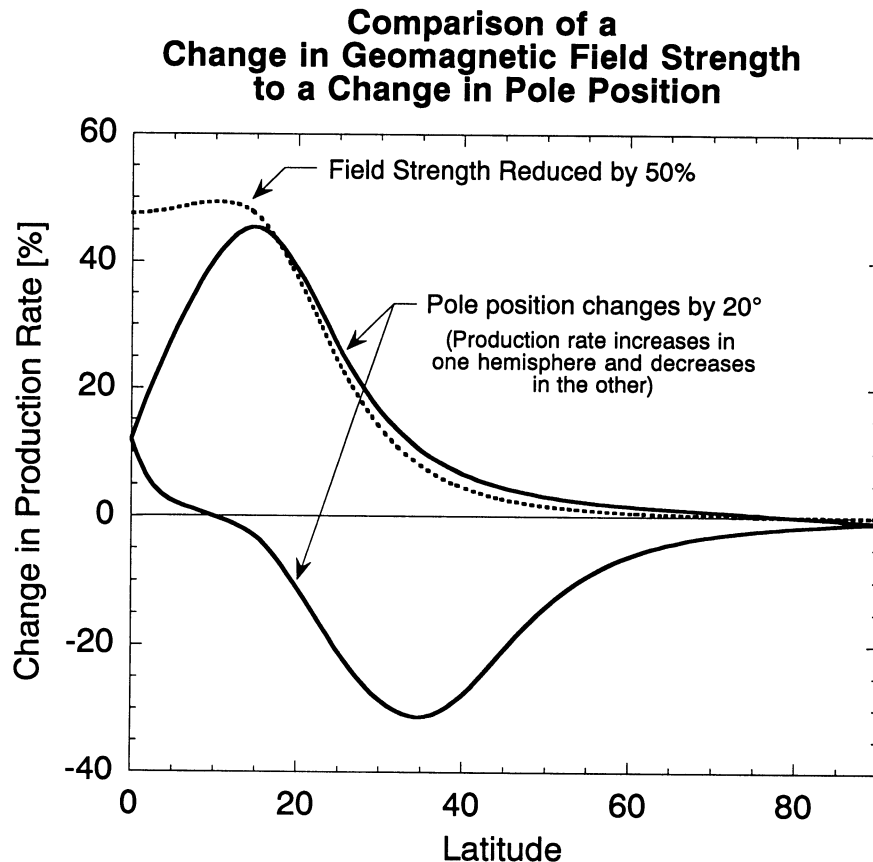
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Production rates of cosmogenic nuclides on Earth undergo temporal variations. These variations are caused by extraterrestrial factors that are expected to affect production rates uniformly everywhere on Earth and by terrestrial factors that cause changes in production rates that vary with location. Accurate age determinations using techniques that rely on the accumulation of cosmogenic nuclides require a knowledge of production rates for the periods during which the samples were exposed. Measurements are now made with sufficient precision that the uncertainty of an age determination is often set by limitations in our knowledge of time-varying production rates. Knowing what causes the changes in production rates is important when transferring production rates measured in one location to other places on earth. Additionally, an appreciation of the possible causes of variations in production rates should inform the selection of locations chosen for calibrations.

For example, the terrestrial factor that has been blithely assumed to have the greatest effect on production rates is the strength of the Earth's magnetic field. However, the figure on the following page shows that at latitudes above  $20^\circ$  an equally important factor may be the position of the magnetic pole relative to the geographic pole. The distinction between field strength and pole position is an important one, not only because different latitudinal profiles must be used to calculate production rates from known changes in field strength and pole position, but also because additional data are needed to determine the position of the pole at the site used for calibration. These additional data may be magnetic declination and inclination measurements at the sampling location, or measurements of relative production rates at positions of similar age and latitude but at different longitude. A recognition of the importance of pole position changes the approach one uses to establish the calibration data.

Other factors that will be addressed include local changes in atmospheric density resulting from sea-level lowering and terrestrial-ice buildup during periods of glaciation and long-term effects of isostatic compensation.



## MONTE CARLO SIMULATION OF *IN-SITU*-PRODUCED COSMOGENIC NUCLIDES

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Cosmogenic nuclides produced *in situ* in terrestrial surface samples provide an important tool for dating and determining erosional histories of landforms. Reliable interpretation of the measured nuclide contents requires a good understanding of fundamentals of nuclide production mechanisms. We present a pure physical model (Masarik and Reedy 1995) for the simulation of the relevant processes enabling an investigation of nuclide production dependence on depth, composition, atmospheric thickness, and geomagnetic field intensity.

In our simulation model (Masarik and Reedy 1994, 1995), the spectra of particles inducing reactions that produce cosmogenic nuclides are calculated by Monte Carlo numerical simulations using the LAHET Code System (LCS) and the GEANT code. Having calculated neutron fluxes with these codes, the production rates of nuclides are determined by integrating over energy the product of these fluxes with experimental and evaluated cross sections for the reactions producing each nuclide. Technical details of the LCS model and our approach are described in Masarik and Reedy (1995). This approach is similar to that often used and well tested by us for cosmogenic-nuclide production rates in lunar samples and meteorites (*e.g.*, Masarik and Reedy 1994).

We simulated the irradiation of Earth with an isotropic GCR particle flux with an energy distribution corresponding to the GCR primary particle flux averaged over a solar cycle. The Earth was modeled as a sphere with a 6378-km radius and an average crustal elemental composition. The model atmosphere has a 1030-g/cm<sup>2</sup> thickness and its standard composition, density, and temperature profiles (Masarik and Reedy 1995). Running 10<sup>7</sup> primary particles, we obtained surface neutron fluxes with statistical errors of ~8%. Although we calculated muon fluxes, their contribution to the production rate was not considered by us because of the lack of the data needed to convert muon fluxes to production rates.

Except for H and Fe, most changes in the surface composition or the addition of other elements to the assumed composition have very little effect on the calculated fluxes (Masarik and Reedy 1994). Therefore elemental production rates valid for many types of rocks can be obtained from these fluxes. The total production rates (in atoms per gram-element per year) only by neutrons and for high latitudes and sea level are given by

$$P(^{10}\text{Be}) = 10.87[\text{O}] + 0.52[\text{Mg}] + 0.39[\text{Si}] + 0.45[\text{Al}] + 0.16[\text{Fe}]$$

$$P(^{14}\text{C}) = 31.3[\text{O}] + 5.3[\text{Mg}] + 4.2[\text{Al}] + 4.3[\text{Si}] + 1.2[\text{Fe}]$$

$$P(^{26}\text{Al}) = 225[\text{Al}] + 77[\text{Si}] + 0.15[\text{Fe}]$$

$$P(^{36}\text{Cl}) = 129[\text{K}] + 65[\text{Ca}] + 16[\text{Ti}] + 0.9[\text{Fe}]$$

$$P(^{21}\text{Ne}) = 98[\text{Na}] + 131[\text{Mg}] + 65[\text{Al}] + 39[\text{Si}] + 4[\text{Ca}] + 0.20[\text{Fe}]$$

$$P(^3\text{He}) = 135[\text{O}] + 116[\text{Mg}] + 107[\text{Al}] + 111[\text{Si}] + 61[\text{Ca}] + 40[\text{Fe}]$$



in which the target-element concentrations, such as [O], are in weight fractions. The calculated rates for most minor target elements are not well tested and could have considerable uncertainties. Latitude-altitude production rates for all above listed nuclides were also calculated. The results of our model calculations are in good agreement with most recent measurements (Masarik and Reedy 1995).

## **PRODUCTION RATE OF $^{10}\text{Be}$ AND $^{26}\text{Al}$ ON THE SURFACE OF THE EARTH AND UNDERGROUND**

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To apply *in-situ* produced cosmogenic nuclides to the study of Earth surface processes, production rates at the surface of the Earth and the attenuation length of the production underground must be known. I review present understanding of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  production rates from quartz, based on different methods and covering various time scales. Each method has its merits and limitations that must be understood in order to assess the reliability of the production rates.

A. *Geological Methods*: Geological methods require that the exposure conditions of the samples studied be constant and that they be well known. Production rates are averages over changes of the Earth's magnetic field and of the primary cosmic ray flux.

1. Concentration of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in quartz from glacially polished rocks (Nishiizumi *et al.* 1989): Since samples are uneroded, nuclides with various half-lives can be compared in the same sample (Neidermann *et al.* 1994). Uncertainties in the exposure age (glaciation age) and in the geomagnetic latitude correction must be considered.
2. Concentration of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in saturated rocks (Nishiizumi *et al.* 1991; Brown *et al.* 1991): The high concentrations of cosmogenic  $^{21}\text{Ne}$  (Hudson *et al.* 1991) insure that the activities of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in certain Antarctic rocks are saturated (decay rate is the same as the production rate). Effects of changing geomagnetic intensity can be ignored for these high latitude samples. Because of the possible influence of erosion, the production rate estimates are lower limits. In addition, there is an uncertainty arising from uplift of the samples during the million year exposure period.

B. *Theoretical Calculation* (ex. Masarik and Reedy 1995): The accuracy of calculated production rates depends on the model and on knowledge of excitation functions. The increasing availability of laboratory data (Imamura *et al.* 1990; Reedy *et al.* 1994) improves the reliability of the results. The method can be applied to samples of various size and shape.

C. *Direct Measurements of  $^{10}\text{Be}$  Production in a Water Target* (Nishiizumi *et al.* 1995): Exposure conditions are unambiguous but the exposure age is very short compared to the geological time scale. There is an uncertainty arising from secular variations of the Earth's magnetic field and of the primary cosmic ray flux.

TABLE 1. Comparison of  $^{10}\text{Be}$  Production Rates (atom/gSiO<sub>2</sub>/yr at sea level, >50°)

Method	Time scale	$^{10}\text{Be}$ production rate (atom/gSiO <sub>2</sub> /yr)	Reference
Water targets	4 solar cycles	5.32 ± 0.27	Nishiizumi <i>et al.</i> 1995
Water targets (after geomagnetic correction)	11,000 yr	6.1 ± 0.3	Nishiizumi <i>et al.</i> 1995
Glacially polished rocks at Sierra Nevada	11,000 yr	6.03	Nishiizumi <i>et al.</i> 1989
Antarctic Rocks	≥ 4 My	6.13	Nishiizumi <i>et al.</i> 1991
Antarctic Rocks	≤ 2.5 My	6.4	Brown <i>et al.</i> 1991
Theoretical calculation	> several My	5.97	Masarik and Reedy 1995

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## COSMIC-RAY NEUTRONS AT GROUND LEVEL: TEMPORAL AND GEODETIC VARIATIONS

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Ground-level cosmic-ray neutrons are the major producer of near-surface cosmogenic isotopes. The inventory of a cosmogenic isotope at a particular location at a particular time depends on the past temporal variation of the neutron flux at that location. To obtain the sensitivity of terrestrial ground-level neutron fluxes on time and space, ground-level fluxes have been calculated using LUIN94 (O'Brien 1995) as a function of geodetic coordinates, altitude, solar activity and magnetic field intensity. The geomagnetic field was treated in considerable detail, using the 1980 Epoch vertical cutoff calculations of Shea and Smart (1983), obtaining the non-vertical cutoffs by means of the approach of Rösler (personal communication) and Heinrich and Spill (1979). Solar modulation was treated in the heliocentric potential approximation. Results are presented graphically. The tabular data underlying these data or the codes used to generate them will be given to anyone interested.

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## **MORE REVEALING HISTORIES OF EXPOSURE: THE RAT URINE STORY**

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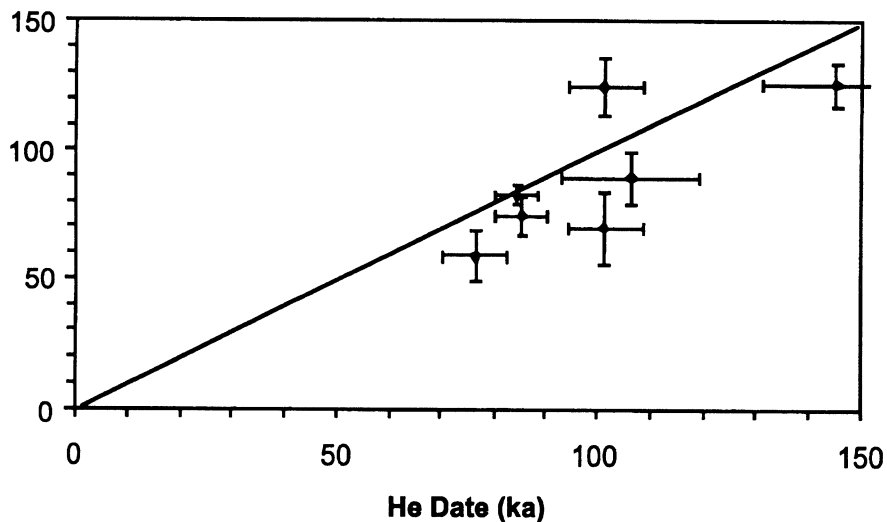
Accurate surface exposure dating using cosmogenic nuclides will require correcting for the effects of secular variations in the production rates of cosmogenic nuclides, presumably mostly due to fluctuations in the intensity of the geomagnetic field. The two main sources of data about these variations are paleomagnetic intensity reconstructions (mostly from sediment cores) and documented discrepancies between  $^{14}\text{C}$  ages and independent ages. Unfortunately, environmental conditions during deposition can affect the accuracy of paleomagnetic intensity reconstructions, and their reliability is difficult to assess. Variations in cosmogenic production can be back-calculated from the  $^{14}\text{C}$  discrepancies, but the magnitude of these discrepancies is strongly affected by changes in oceanic circulation. We have measured  $^{36}\text{Cl}$  in fossil packrat urine in order to provide an independent basis for reconstructing variations in cosmogenic nuclide production. The record of  $^{36}\text{Cl}$  fluctuations thus obtained shares many features with the  $^{14}\text{C}$  and paleomagnetic reconstructions, but indicates a larger and more abrupt decrease in cosmogenic production at the end of the last glacial period.

## COMPARISON OF DATES FOR YOUNG BASALTS FROM THE $^{40}\text{Ar}/^{39}\text{Ar}$ AND COSMOGENIC HELIUM TECHNIQUES

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We have  $^3\text{He}$  surface exposure dates and  $^{40}\text{Ar}/^{39}\text{Ar}$  dates for a number of lavas from three volcanic fields: Potrillo (New Mexico), Lathrop Wells (Nevada), and San Quintin (Baja California, Mexico). The dates span from 20 ka to 150 ka. While agreement among the two types of dates is encouraging, it is not universal. Both types of dates have their associated assumptions that can bias the results: He dates for older lavas may be lower than the eruption age due to erosion, while  $^{40}\text{Ar}/^{39}\text{Ar}$  dates at low radiogenic yields are sensitive to assumptions about the trapped component. A particularly encouraging approach is  $^{40}\text{Ar}/^{39}\text{Ar}$  dating of sanidine separated from xenoliths. At Lathrop Wells, a subset of these xenoliths yields an upper limit for the age of the Q12 flow of  $75 (\pm 2)$  ka, slightly younger than the  $^3\text{He}$  date ( $85 \pm 5$  ka). This observation, as well as the general comparison of Ar and  $^3\text{He}$  dates (figure below), suggests that the integrated production rate for  $^3\text{He}$  is slightly higher at *ca.* 80 ka, compared to that derived from Tabernacle Hill at 17 ka.

### Comparison of He and Ar Dates



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**PRODUCTION SYSTEMATICS OF COSMOGENIC NUCLIDES IN THE EARTH***ROBERT C. REEDY*

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The high-energy (GeV) particles in the galactic cosmic rays (GCR) produce nuclides deep in any object exposed to them. Cosmogenic nuclides have been well studied since the 1950s in meteorites and lunar samples. In extraterrestrial matter, several approaches have been used to determine cosmogenic-nuclide production systematics. The work being done for terrestrial cosmogenic nuclides (*e.g.*, Reedy *et al.* 1994a) follows the approach used for extraterrestrial nuclides.

For extraterrestrial cosmogenic nuclides, initial studies used activities of radionuclides to infer production rates. (For a long, constant exposure to cosmic rays, a radionuclide's decay rate approaches equilibrium with its production rate.) Irradiations of thick targets at high-energy accelerators were done to experimentally simulate GCR irradiations in space and helped in understanding the nuclear processes involved. Accelerators were also used with thin targets to measure cross sections for making these cosmogenic nuclides. Several models were developed for predicting the systematics of the production of cosmogenic nuclides, mainly semi-empirical fits. Recently, numerical simulations based only on physics have been used to study nuclear reactions and allow us to examine the details of how GCR particles interact with matter (Masarik and Reedy, this issue).

The terrestrial case is more complicated because of the Earth's atmosphere and strong geomagnetic field. Most GCR particles interact in the Earth's atmosphere, making mainly  $^{14}\text{C}$ . The Earth's thick (1033 g/cm<sup>2</sup>) atmosphere attenuates the flux of particles reaching the surface by several orders of magnitude. The large difference in the composition and density of the atmosphere compared to extraterrestrial matter makes it difficult to use data from extraterrestrial matter for nuclides made deep in the atmosphere. One major difference is that most pions made in dense extraterrestrial matter interact before they can decay, while most pions in the thin atmosphere decay to muons. These muons are weakly-interacting particles and can go much further than strong-interacting particles such as neutrons and pions. Below a few meters in the Earth's surface, muons are a major source of cosmogenic nuclides. Another complication is that the production rates of a nuclide in the Earth's surface can vary much with the sample's elevation and the location's geomagnetic latitude.

Most production rates of nuclides made in the Earth are now based on a few measurements made with natural samples. Generally, the production rate of a cosmogenic nuclide is converted from that determined at the location's elevation and geomagnetic latitude to the rate at sea level and a high geomagnetic latitude (>60°) with no cutoff of GCR particles. Most measurements have been done on material with a simple composition, often quartz (SiO<sub>2</sub>) or calcite (CaCO<sub>3</sub>).

Several studies have been done at accelerators on production systematics of terrestrial cosmogenic nuclides. Some irradiations were done at the Los Alamos Meson Physics Facility, usually using secondary neutrons near a beam-stop. These irradiations have yielded some production ratios, such as  $^{10}\text{Be}/^{14}\text{C}/^{26}\text{Al}$  in SiO<sub>2</sub> (Reedy *et al.* 1994b), and could be used to get relative production rates from pure elements. Some accelerators are also sources of muon beams. Work is also being done on measuring yields of nuclides from fast muons or muons stopping in various targets (Reedy *et al.* 1994c).

As the ratio of neutrons to protons in the Earth's surface is higher than in most meteorites and lunar samples, cross sections for the production of terrestrial cosmogenic nuclides by energetic neutrons are very important. However, only a few neutron cross sections have been measured for the nuclides

of interest, mainly at energies below 20 MeV. More cross sections are needed for terrestrial cosmogenic nuclides, especially for neutron energies above 20 MeV.

Neutron-transport codes, such as the Monte Carlo N-Particle (MCNP) code, can be used to study nuclides made by neutron-capture reactions and have shown that rates for the  $^{35}\text{Cl}(n,\gamma)^{36}\text{Cl}$  reaction depend on the rock's composition and water content. The LAHET Code System (LCS) and similar codes have been used very successfully for studying cosmogenic nuclides in extraterrestrial matter. LCS was recently used to study production of nuclides in the Earth (Masarik and Reedy, this workshop).

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### WORKSHOP ON SECULAR VARIATIONS IN THE RATES OF PRODUCTION OF COSMOGENIC NUCLIDES ON EARTH: PALEOMAGNETIC AVERAGES OF GEOMAGNETIC LATITUDE

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The production rate of cosmogenic nuclides depends on the geomagnetic cutoff rigidity, which in turn depends upon geomagnetic latitude. The geomagnetic dipole moment axis moves over time, one of the characteristic patterns of secular variation. Over the past 300 yr, the latitude of the northerly end of the dipole axis has decreased from *ca.* 83°N to 78°N, while longitude has drifted westward from 320°E to 290°E (Merrill and McElhinny 1983). On the other hand, a critical assumption of paleomagnetism is the geocentric axial dipole field hypothesis, whereby the magnetic direction at any locality will average over several millennia to that of an axial geocentric dipole.

Data of Ohno and Hamano (1992) were used to model the effect of dipole secular variation on average geomagnetic latitude. This compilation gives average pole positions at 500-yr increments for the past 10,000 yr, based on archaeomagnetic and sediment paleomagnetic data. For several sites on the Earth's surface, the average pole position was integrated back in time from the present to 10,000 yr ago. Differences between geomagnetic and geographic latitudes (here called the latitude difference) were calculated for the time-integrated average pole positions at 500-yr increments. Averaging from the present back to 1–4 ka gives a median difference of 1.7°; from 4–7 ka, it is 1.4°; from 7–10 ka, it is also 1.4°. A similar analysis was carried out using pole positions of Merrill and McElhinny (1983: Table 4.1) at 100-yr increments back to 2000 yr BP. Averaging from the present back to 200–800 BP gives a median difference of 4.2°. Thus, for exposure times longer than several centuries, the latitude error is typically 1.5°.

The Laschamp excursion, involving anomalous paleomagnetic directions and intensities, has been observed in some but not all data sets between 30 and 50 ka (Thouveny and Creer 1992). If such an excursion were due to a tilt of the dipole axis, its effect on average geomagnetic latitude could also be calculated. If the pole moves linearly to a latitude of 45° and back during a 5-ka long excursion (ignoring changes in dipole moment), the integrated average latitude difference is down to 2° after 50,000 yr; for a short 200-yr excursion (Thouveny and Creer 1992), the difference is down to 2° after 200 yr.

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## COSMOGENIC CHLORINE-36 PRODUCTION RATES FROM CALCIUM AND POTASSIUM

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Advantages of exposure dating with cosmogenic Chlorine-36 are high <sup>36</sup>Cl production rates in a number of common minerals, and the high sensitivity of AMS Cl isotopic analyses. A disadvantage is the variety of <sup>36</sup>Cl-producing reactions—spallation reactions on K and Ca, muon capture reactions on these elements, and capture (by <sup>35</sup>Cl) of secondary neutrons produced in spallation, muon capture and fast muon interactions. Complications arise through the different latitude, altitude, depth and compositional dependences of these reactions. Also, errors in calibration of any one reaction tend to propagate into calibration of the others. In estimating <sup>36</sup>Cl production rates from geological surfaces, our strategy has been to: 1) work with separated minerals—calcite, Ca-feldspar and K-feldspar—to maximize target element abundances (enhancing analytical sensitivity) and Ca/Cl and K/Cl ratios (enhancing contributions from spallation relative to slow neutron capture); 2) use high altitude sites for calibrating spallation reactions and deep sub-surface samples for calibrating muon reactions; and 3) suppress <sup>36</sup>Cl measurement errors and reduce dependence on accurate Cl analyses by adding Cl carrier to samples.

Results are summarized below. Quoted uncertainties are  $\pm 1 \sigma$  (68% confidence) based on analytical errors and indicated uncertainties in calibration surface ages. Uncertainties refer to production rates at the calibration sites; additional errors in scaling to sea-level and high latitude have not been propagated here. Spallation and muon reactions have been scaled separately using the procedures of Lal (1991: EPSL 104, 424). Latitude corrections are based on effective geomagnetic latitudes ( $\lambda_{\text{eff}}$ ) of the calibration sites over their exposure times. Sidereal ages are used for calibration, to avoid errors due to non-linearity in the radiocarbon timescale.

1. *Ca Spallation*: Our primary calibration is based on Ca-feldspar from the Tabernacle Hill basalt ( $t_{\text{exp}} = 17.3 \pm 0.5$  ka), which gives a Ca spallation rate of  $152 \pm 5$  atom (g Ca)<sup>-1</sup> a<sup>-1</sup> at 1445 m,  $\lambda_{\text{eff}} = 40.9^\circ$ . Five measurements were made on feldspar from three separate rock samples. Ca-spallation accounts for 86–88% of <sup>36</sup>Cl in the samples, with 5% due to <sup>40</sup>Ca( $\mu^-$ ,  $\alpha$ )<sup>36</sup>Cl, 7% due to K reactions and 1–2% due to neutron capture. The equivalent rate at sea level and high latitude is  $48.8 \pm 1.7$  atom (g Ca)<sup>-1</sup> a<sup>-1</sup>.
2. *Muon Reactions in Calcite*: <sup>36</sup>Cl measurements on a 20-m profile in calcite marble at Wombeyan in SE Australia (620 m,  $\lambda = 34.3^\circ$ ) give a <sup>40</sup>Ca( $\mu^-$ ,  $\alpha$ )<sup>36</sup>Cl production rate of  $0.011 \pm 0.0013$  atom <sup>36</sup>Cl per stopped negative muon and a secondary neutron yield of  $0.41 \pm 0.1$  neutrons per stopped negative muon in calcite. Assuming a negative muon stopping rate of  $175 \mu^- \text{ g}^{-1} \text{ a}^{-1}$  at sea-level and high latitude, the surface production rate for the <sup>40</sup>Ca ( $\mu^-$ ,  $\alpha$ )<sup>36</sup>Cl reaction is  $4.8 \pm 0.5$  atom (g Ca)<sup>-1</sup> a<sup>-1</sup> (i.e., 9% of total production from Ca). The natural calibration avoids the need to know values for muon reaction parameters such as the chemical compound factor and  $\alpha$ -emission probability. Estimates of these factors are required to apply the results to

minerals other than calcite, however. Assuming a Fermi-Teller (“z-law”) distribution of muon captures between Ca, C and O in calcite, we derive an  $\alpha$ -emission probability following muon capture of  $0.033 \pm 0.004$ , well within the range of existing measurements and calculations ( $0.004 < P\alpha < 0.15$ ).

3. <sup>36</sup>Cl production from K: So far only surface samples of K-feldspar have been measured, precluding a breakdown of total production from K (PK) into component reactions. Samples from glacial pavements in the Sierra Nevada, Scotland and Antarctica have been measured. Assuming  $t_{\text{exp}}=13.1$  ka, samples from the Sierra Nevada give  $PK=1460 \pm 88$  atom (g K)<sup>-1</sup> a<sup>-1</sup> at 3000 m and  $\lambda_{\text{eff}}=37.4^\circ$ . Concordant results ( $\chi^2/n=0.8$ ,  $n=10$  measurements) are obtained from samples at 4 sites, at elevations between 3000 and 3600 m and with chloride concentrations from 10–310 ppm. Reactions on K account for 76–99% of production in the samples. Consistency between samples with low and high Cl contents is obtained assuming a surface neutron capture rate of  $2450$  n g<sup>-1</sup> a<sup>-1</sup> at 3000 m and  $37.4^\circ$ , close to the scaled value of Zreda *et al.* (1991: EPSL 105: 94–109). Assuming 5% of K production at sea level is due to muon capture, these data give  $PK = 180 \pm 11$  atom (g K)<sup>-1</sup> a<sup>-1</sup> at sea level and high latitude. Samples from pavements exposed by retreat of Loch Lomond (Y. Dryas) Stage ice ( $t_{\text{exp}} = 11.5 \pm 0.3$  ka) in Scotland give  $PK = 313 \pm 25$  atom (g K)<sup>-1</sup> a<sup>-1</sup> at 520 m and  $\lambda = 58.5^\circ$ . Neutron capture corrections are 18–22% for these samples. Again assuming 5% of total K production due to muon capture, these data give  $PK = 189 \pm 15$  atom (g K)<sup>-1</sup> a<sup>-1</sup> at sea level and high latitude, in excellent agreement with results from the Sierra Nevada. Two samples of K-feldspar from the Trans-Antarctic Mountains ( $\lambda = 77.5^\circ$ ), apparently saturated with <sup>36</sup>Cl, give production rates of  $1350 \pm 50$  atom (g K)<sup>-1</sup> a<sup>-1</sup> at 2050 m and  $1230 \pm 40$  atom (g K)<sup>-1</sup> a<sup>-1</sup> at 2000 m, with negligible (<2%) corrections for neutron capture on <sup>35</sup>Cl. Equivalent rates at sea level, scaled as above, are  $PK = 239 \pm 10$  and  $227 \pm 8$  atom (g K)<sup>-1</sup> a<sup>-1</sup>. The samples have been measured repeatedly to confirm that the 25% discrepancy with the scaled values from Scotland and the Sierra Nevada is real and must be accounted for either by the scaling procedure, or secular variation in the cosmic ray flux.

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## TEMPORAL VARIATIONS IN GLOBAL ATMOSPHERIC <sup>14</sup>C AND ITS PRODUCTION

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The atmospheric radiocarbon record of the past 20,000 yr is an integrated response to climatic, solar and geomagnetic forcing. The time dependencies of the forcing functions play an important role in shaping the atmospheric <sup>14</sup>C profile. Geomagnetic forcing manifests itself mainly on millennia time scales, whereas solar forcing is more restricted to decadal and century time intervals. Oceanic thermohaline circulation change (and associated climate perturbation) sufficiently large to induce atmospheric <sup>14</sup>C change operates on decadal to millennia time scales.

<sup>14</sup>C age calibration is needed because conventional <sup>14</sup>C dates do not take atmospheric <sup>14</sup>C variability into account. Use of our knowledge of forcing factors in carbon reservoir models will generate “hypothetical” calibration curves, but the limitations of our knowledge, especially of solar and oce-



anic variability, are such that calculated calibration curves lack much of the detail found in measured Holocene tree-ring  $^{14}\text{C}$ .

The record of  $^{14}\text{C}$  change can be extended beyond the interval covered by tree rings by using  $^{14}\text{C}$  and U/Th dated corals. However, the marine surface waters greatly attenuate production-rate induced  $^{14}\text{C}$  variations, and thus the calibration curve extension loses much century-type production rate detail.

## **DETERMINATION OF $^{36}\text{Cl}$ PRODUCTION RATES FROM THE DEGLACIATION HISTORY OF WHIDBEY AND FIDALGO ISLANDS, WASHINGTON**

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Cosmogenic  $^{36}\text{Cl}$  is produced in surface rocks by thermal neutron activation of  $^{35}\text{Cl}$ , spallation of  $^{39}\text{K}$  and  $^{40}\text{Ca}$  and slow negative muon capture by  $^{40}\text{Ca}$ . Production parameters for the  $^{36}\text{Cl}$  isotopic system were refined using the well-constrained deglaciation history of Whidbey and Fidalgo Islands, Washington, USA. Calculated production rates due to spallation are  $1770 \pm 121$  atoms  $^{36}\text{Cl}$  ( $\text{kg}^{-1}$  rock) ( $\text{percent K}_2\text{O}^{-1}$ )  $\text{yr}^{-1}$  and  $641 \pm 20$  atoms  $^{36}\text{Cl}$  ( $\text{kg}^{-1}$  rock) ( $\text{percent CaO}^{-1}$ )  $\text{yr}^{-1}$ . The calculated effective thermal neutron absorption rate is  $(3.53 \pm 0.07) \times 10^5$  neutrons  $\text{kg}^{-1}$  of rock  $\text{yr}^{-1}$ . Production due to muon capture by  $^{40}\text{Ca}$  was estimated using previously calculated values scaled to high geomagnetic latitude and sea level.

Consistency between calculated  $^{36}\text{Cl}$  ages using the production rates reported here, and independent  $^{14}\text{C}$  ages that constrain the timing of deglaciation of the Cordilleran Ice Sheet over three disparate locations, substantiates the validity of the  $^{36}\text{Cl}$  production rates for high-latitude ( $>60^\circ$ ) locations, integrated over the last  $\sim 15,200$  cal yr. Evaluation of potential sources of error indicate that uncertainty of AMS and total Cl measurements are the dominant factors controlling the overall uncertainty of the  $^{36}\text{Cl}$  ages. Geologic factors such as surface erosion and/or prior exposure history are also important sources of error that were addressed by this study; however, independent testing of the production rates on independent surfaces of known age suggest that these potential problems were minor for the 64 calibration samples.

Analytical uncertainties of  $\pm 5\text{--}6\%$  ( $1\sigma$ ) for the calibration samples indicate that it should be possible to resolve late-glacial landforms from those of full-glacial age using the  $^{36}\text{Cl}$  dating method, providing the analytical uncertainty of AMS and total Cl measurements is  $<5\%$  and geological problems are accurately constrained. However, without greatly improving the precision of AMS and total Cl measurements, it is unlikely that the  $^{36}\text{Cl}$  dating method will be able to resolve ages of, or correlate landforms associated with, short-term climatic episodes such as the late-glacial Younger Dryas event.

## RECENT ADVANCES IN DETERMINING ABSOLUTE AND RELATIVE PALEOINTENSITY VARIATIONS OF THE GEOMAGNETIC FIELD

*KENNETH L. VEROSUB*

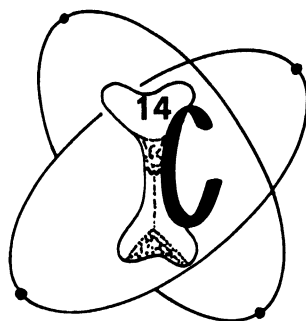
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For the past 30 yr, the focus in paleomagnetism has been on the direction of the paleomagnetic vector rather than on its intensity. The reason for this "two-dimensional" view of a three-dimensional quantity is that determination of the paleointensity is inherently more complex, more difficult, and less reliable than determination of the paleomagnetic direction. This situation may be changing as new techniques and new instrumentation are brought to bear on these problems.

For determinations of the absolute paleointensity of the geomagnetic field, the standard methodology has, for many years, been the Thellier-Thellier double heating experiment. This approach can be used with material that carries a thermal remanent magnetization, such as lava flows, hearths and pottery. Unfortunately, the Thellier-Thellier experiment is very time-consuming, and the percentage of samples that give unsatisfactory results can be quite high. As a result, the database of absolute paleointensity determinations is still relatively small. Recently, several research groups have tried to develop mineral magnetic screening criteria for identifying, in advance, those samples that were likely to produce satisfactory results in the Thellier-Thellier experiment. Although this work promises to increase the success rate for absolute paleointensity determinations, it does not address another important problem, namely, the available material seldom represents a continuous sampling of the geomagnetic field through time. This problem is particularly acute for studies involving lava flows but it can also arise when archaeological materials are involved.

Sediments, on the other hand, offer the possibility of a continuous record of geomagnetic intensity variations. For sediments, however, the intensity of magnetization depends on the intensity of the geomagnetic field at the time the magnetization was acquired and on several other factors, including the concentration and grain size of the magnetic carriers. Many years ago it was proposed that the relative intensity of the geomagnetic field intensity could be determined with a normalization based on the intensity of a laboratory-induced magnetization. It was soon recognized that too much variation in the size or concentration of the magnetic grains could be a problem, and various selection criteria were proposed. In recent years, a globally consistent picture of relative paleointensity variations is beginning to emerge, at least for the last 250,000 yr. In addition, there has been the intriguing suggestion that variations in relative paleointensity over the past four million years are linked to polarity transitions. However, there is still disagreement about the interpretation of significant spatial and temporal variations that are seen in relative paleointensity records over both time scales. In addition, it is not clear whether the absolute and relative paleointensity records are mutually consistent.

With regard to the determination of production rates of cosmogenic nuclides, it is important to recognize that both absolute and relative paleointensity determinations are not easy to make nor simple to interpret, and that the inherent problems and complexities could lead to problems in calculations based upon them. On the other hand, the most important observation may be that geomagnetic intensities during the past 10,000 yr have been somewhat higher than the average for the past 250,000 yr. If this conclusion is correct, then temporal and spatial variations that are present over the longer time interval could have had a relatively minor influence on production rates.



## 3rd INTERNATIONAL SYMPOSIUM

# ***<sup>14</sup>C and ARCHAEOLOGY***

LYON, FRANCE

**6–10 APRIL 1998**

The 3rd International Symposium <sup>14</sup>C and Archaeology will be held in Lyon, France, at Claude-Bernard University, from Monday, April 6, through Friday, April 10, 1998.

This conference is intended to allow archaeologists and <sup>14</sup>C specialists to meet and interact. It is especially meant to draw up chronological syntheses based upon <sup>14</sup>C dates, while also reviewing recent advances in radiocarbon technology.

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Vol. 39, No. 1, 1997	September 1, 1996
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*Half-life of  $^{14}\text{C}$ .* In accordance with the decision of the Fifth Radiocarbon Dating Conference, Cambridge, England, 1962, all dates published in this volume (as in previous volumes) are based on the Libby value, 5568 yr, for the half-life. This decision was reaffirmed at the 11th International Radiocarbon Conference in Seattle, Washington, 1982. Because of various uncertainties, when  $^{14}\text{C}$  measurements are expressed as dates in years BP, the accuracy of the dates is limited, and refinements that take some but not all uncertainties into account may be misleading. The mean of three recent determinations of the half-life,  $5730 \pm 40$  yr, (*Nature*, 1962, Vol. 195, No. 4845, p. 984), is regarded as the best value presently available. Published dates in years BP can be converted to this basis by multiplying them by 1.03.

*AD/BC Dates.* In accordance with the decision of the Ninth International Radiocarbon Conference, Los Angeles and San Diego, California, 1976, the designation of AD/BC, obtained by subtracting AD 1950 from conventional BP determinations is discontinued in *RADIOCARBON*. Authors or submitters may include calendar estimates as a comment, and report these estimates as cal AD/BC, citing the specific calibration curve used to obtain the estimate. Calibrated dates should be reported as "cal BP" or "cal AD/BC" according to the consensus of the Twelfth International Radiocarbon Conference, Trondheim, Norway, 1985.

*Measuring  $^{14}\text{C}$ .* In Volume 3, 1961, we endorsed the notation  $\Delta$ , (Lamont VIII, 1961), for geochemical measurements of  $^{14}\text{C}$  activity, corrected for isotopic fractionation in samples and in the NBS oxalic-acid standard. The value of  $\delta^{14}\text{C}$  that entered the calculation of  $\Delta$  was defined by reference to Lamont VI, 1959, and was corrected for age. This fact has been lost sight of, by editors as well as by authors, and recent papers have used  $\delta^{14}\text{C}$  as the observed deviation from the standard. At the New Zealand Radiocarbon Dating Conference it was recommended to use  $\delta^{14}\text{C}$  only for age-corrected samples. Without an age correction, the value should then be reported as percent of modern relative to 0.95 NBS oxalic acid (Proceedings of the 8th Conference on Radiocarbon Dating, Wellington, New Zealand, 1972). The Ninth International Radiocarbon Conference, Los Angeles and San Diego, California, 1976, recommended that the reference standard, 0.95 NBS oxalic acid activity, be normalized to  $\delta^{13}\text{C} = -19\text{‰}$ .

In several fields, however, age corrections are not possible.  $\delta^{14}\text{C}$  and  $\Delta$ , uncorrected for age, have been used extensively in oceanography, and are an integral part of models and theories. Thus, for the present, we continue the editorial policy of using  $\Delta$  notations for samples not corrected for age.

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