# Continental weathering as a driver of Late Cretaceous cooling: new insights from clay mineralogy of Campanian sediments from the southern Tethyan margin to the Boreal realm

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## Abstract

New clay mineralogical analyses have been performed on Campanian sediments from the Tethyan and Boreal realms along a palaeolatitudinal transect from 45° to 20°N (Danish Basin, North Sea, Paris Basin, Mons Basin, Aquitaine Basin, Umbria-Marche Basin and

Tunisian Atlas). Significant terrigenous inputs are evidenced by increasing proportions of detrital clay minerals such as illite, kaolinite and chlorite at various levels in the mid- to upper Campanian, while smectitic minerals predominate and represented the background of the Late Cretaceous clay sedimentation. Our new results highlight a distinct latitudinal distribution of clay minerals, with the occurrence of kaolinite in southern sections and an almost total absence of this mineral in northern areas. This latitudinal trend points to an at least partial climatic control on clay mineral sedimentation, with a humid zone developed between 20° and 35°N. The association and co-evolution of illite, chlorite and kaolinite in most sections suggest a reworking of these minerals from basement rocks weathered by hydrolysis, which we link to the formation of relief around the Tethys due to compression associated with incipient Tethyan closure. Diachronism in the occurrence of detrital minerals between sections, with detrital input starting earlier during the Santonian in the south than in the north, highlights the northward progression of the deformation related to the anticlockwise rotation of Africa. Increasing continental weathering and erosion, evidenced by our clay mineralogical data through the Campanian, may have resulted in enhanced $CO_2$ consumption by silicate weathering, thereby contributing to Late Cretaceous climatic cooling.

Keywords: Campanian, Late Cretaceous cooling, clay minerals, carbon isotope stratigraphy, climatic belt, continental weathering

## 1. Introduction

The Late Cretaceous is characterised by a long-term global climatic cooling from the Turonian onward, with a marked acceleration during the Campanian (Huber et al., 1995; Pucéat et al., 2003; Friedrich et al., 2012; Linnert et al., 2014). This period shows evidence of an overall decrease in atmospheric $CO_2$ levels that likely contributed to this global cooling, although the data remain scarce and do not allow to identify the timing and phases of $CO_2$ decline within the Late Cretaceous (Royer et al., 2012; Wang et al., 2014; Franks et al., 2014). Decreasing mantle degassing linked to variations in seafloor production rates and continental arc magmatism has been invoked to explain the observed decline in $CO_2$ levels during the Late Cretaceous (Berner, 2004; Cogné and Hummler, 2006; Van Der Meer et al., 2014; McKenzie et al., 2016).

Continental silicate weathering also governs atmospheric $CO_2$ on a multi-million year time scale (Berner, 1990, 2004; Raymo and Ruddiman 1992; Dessert et al., 2003), but this process remains poorly explored for the Late Cretaceous. Yet this period was marked by major geodynamic changes, which included the initiation of Tethys Ocean closure (Dercourt et al., 1986). This event, which is linked to the convergence of Africa toward Eurasia, was associated with the development of topographic relief around the Tethys, including lithospheric folds in Morocco (Frizon de Lamotte et al., 2011), and the 5000 km long chain of relief developed across North Africa and the Middle East by the Ayyubid orogeny (Şengör and Stock, 2014). This increased relief might be expected to have induced an increase in continental silicate weathering, enhancing $CO_2$ consumption.

Clay minerals assemblages may be used to assess fluctuations in continental weathering intensity (Chamley, 1989; Hermoso and Pellenard, 2014). Numerous clay minerals data exist for the Cenomanian, Turonian and Maastrichtian stages but they remain scarce for the Santonian–Campanian interval although many significant environmental changes occurred at that time. A major reorganization of oceanic circulation took place during the Santonian–Campanian, evidenced by neodymium isotope data (Robinson et al., 2010; Martin et al., 2012; Moiroud et al., 2016). The Campanian stage was also characterised by a significant cooling, as indicated by $\delta^{18}O$ values of benthic foraminifera and $TEX_{86}$ data (Friedrich et al., 2012; Linnert et al., 2014; O'Brien et al., 2017).

At a regional scale, preliminary clay minerals data from the Tercis-les-Bains section (Aquitaine basin, North Atlantic influenced) and from the Poigny borehole (Paris basin, Boreal influenced) have shown increasing inputs of detrital illite and kaolinite during the Campanian (Chenot et al., 2016), coinciding with a global carbon-isotope negative excursion, the so-called "Late Campanian Event" of Jarvis et al. (2002). This suggests that changes in the carbon cycle at this time were accompanied by an increase of continental weathering. In order to explore the spatial and temporal extent of these modifications in continental weathering, we have acquired new data for clay minerals assemblages from Campanian sediments through 6 different sections and boreholes from the Tethyan and Boreal realms, along a transect from ~18° to ~42°N palaeolatitude. Combined with previously published data sets, our work provides the first constrains at the Tethyan scale on variations in continental weathering induced by tectonic uplift during the Campanian.

## 2. Geodynamic framework and global palaeogeography of the studied sites

From the mid-Cretaceous onward, global plate tectonic changes induced modification of the tectonic stress field in Europe. The Tethys Ocean began to close due to the anticlockwise movement of Africa, with: (1) at the northern margin, the opening of the Bay of Biscay and active subduction zones in Apulia, the Dinarides and Hellenids; and (2) at the southern margin, the development of an intra-oceanic orogenic belt (Smith, 1971; Dewey et al., 1973; Charvet, 1978; Bárdossy and Dercourt., 1990; Faccenna et al., 2001; Blakey, 2008; Kley and Voigt, 2008; Voigt et al., 2008; Fig. 1).

To the north, the interplay of extensional and compressional tectonics, resulting from the west-central Europe's thin lithosphere pinch between Baltica's and Africa's cratonic lithospheres, induced NW–SE striking thrust faulting on the European plate (Kley and Voigt, 2008). These processes caused the development of subsiding basins (e.g. Sorgenfrei-Tornquist Zone), and inversions of former depocentres (e.g. Mid-Polish Trough), with different rates of subsidence (Kley and Voigt, 2008; Voigt et al., 2008). These newly created reliefs provided detrital particles into the adjacent seas (Fig. 1). The geology of the southern margin of Central Europe resulted from the convergence linked to the subduction zone between the European and African plates.

Campanian palaeogeographic reconstructions show that the northern Tethyan margin was partly covered by epicontinental seas (Fig. 1), with emerged lands representing remnants of Variscan relief (e.g. Armorican, Central, Iberian, Ebro, Welsh, Rhenish, Bohemian massifs), the inverted Mid-Polish Anticline (Voigt et al., 2008), and regional shoals (Dalmatian shoal, High Karst; Charvet, 1978). The southern passive continental margin of the Tethys Ocean was a wide platform (e.g. Saharan platform, Syrte basin), influenced by several detrital sources, including locally emerged land masses such as Kasserine Island in Tunisia (Kadri et al., 2015) and the north-western African craton (Fig. 1). However, the palaeogeography of Central Europe is more difficult to reconstruct because of the large area affected by erosion during the Late Cretaceous inversion (Voigt et al., 2008; Wolfgring et al., 2016; Neuhuber et al., 2016).

## 2.1. Boreal Realm

### 2.1.1. Danish North Sea and eastern Danish basin: Stevns-2 and Adda-3 boreholes

The Chalk Group of the Danish North Sea is well studied for its hydrocarbons reservoir properties (Hardman, 1982; Megson and Tygesen, 2005). During the Late Cretaceous, the Danish basin was bordered by the Baltic Shield to the northeast, the Grampian High to the northwest, and the Rhenish–Bohemian Massif to the south (Fig. 1).

The 350 m long Stevns-2 core was drilled in Boesdal Quarry (55°15'31''N 12°24'04''E) located in the eastern part of the Danish basin (palaeolatitude ~42°N; Philip and Floquet, 2000; Fig. 1). The core recovers a complete succession of upper Campanian to Maastrichtian chalks, with a distinctive interval of alternating chalk-marl in the upper Campanian, a feature that is also observed in the nearby Stevns-1 core and appears to characterise the whole Stevns peninsula (Thibault et al., 2016a). The calcareous nannofossils biostratigraphy, high-resolution carbon- and oxygen-isotope chemostratigraphy, and sedimentology of the Stevns-2 core have been described by Boussaha et al. (2016, 2017; Fig. 2).

The Adda-3 well in the Danish Central Graben (55°47'50"N 04°53'26"E) is located in the southern part of the North Sea rift system (palaeolatitude ~45°N; Philip and Floquet, 2000; Fig. 1). The Campanian interval, composed of bioturbated white chalks with occasional thick flint bands and marly layers, occurs between 2200.8–2260.8 m depth. The calcareous nannofossils biostratigraphy and stable-isotope geochemistry ($\delta^{13}C$, $\delta^{18}O$) have been presented by Perdiou et al. (2016; Fig. 2).

### 2.1.2. Mons Basin: Cbr-7 borehole

The Mons basin (southern Belgium) was a transitional area between the North Sea and the Paris basin to the WSW, bordered by the Rhenish Massif to the ENE (palaeolatitude ~37°N; Philip and Floquet, 2000; Fig. 1). Based on lithology, the Campanian chalk of the Mons basin has been subdivided into three formations (Cornet and Briart, 1870; Briart and Cornet, 1880):

the Trivières Chalk (white to grey marly chalk without flint); the Obourg Chalk (fine white chalk with flint in the north of the Mons basin); and the Nouvelles Chalk (fine white chalk without flint).

The 75 m-deep Cbr-7 borehole was drilled on the northern margin of the Hainaut-Sambre quarry (50°25’10”N 04°1’33”E) located in the southeast of the Mons basin (Fig. 1). Robaszynski and Anciaux (1976) subdivided the succession into the: Trivières Chalk (75.0–46.5 m depth); Obourg Chalk (46.5–30.5 m); and Nouvelles Chalk (30.5–2.4 m depth). Biostratigraphic data are scarce, but the uppermost part of the Trivières Chalk, the Obourg Chalk and Nouvelles Chalk have been attributed to the lower part of the upper Campanian according to the vertical distribution of foraminifera, belemnites and echinoids (Robaszynski and Christensen, 1989; Fig. 3). A hardground exhibiting high Mn concentrations, acquired by inductively coupled plasma-atomic emission spectrometry (ICP-AES) in the original study of Richard et al. (2005) and an important $\delta^{13}C$ negative excursion of 0.5‰ amplitude occurs in the uppermost part of the Trivières Chalk.

## 2.2. Tethyan Realm

### 2.2.1. Umbria-Marche basin: Gubbio – la Bottaccione and Furlo – Upper Road sections

A thick succession of Upper Cretaceous pelagic carbonates was deposited in the Umbria-Marche basin in central Italy. During the Late Cretaceous, this deep basin was surrounded by the High-Karst to the northeast (Charvet, 1978; palaeolatitude ~25°N; Philip and Floquet, 2000; Fig. 1). The symmetric NE–SW anticline of the Gubbio – la Bottaccione section (43°21’45”N 12°34’57”E; Fig. 1) exposes a succession of pelagic carbonates from the Upper Jurassic to the Palaeocene (~400 m-thick), followed by the first terrigenous turbidites within the Miocene (Arthur and Fisher, 1977). The Campanian–Maastrichtian Scaglia Rossa Formation is composed of pelagic carbonates with small quantities of iron oxides, including magnetite and hematite, responsible for the pink colour of the limestones (Lowrie and Alvarez, 1977; Channell et al., 1982; Lowrie and Heller, 1982).

The Campanian Scaglia Rossa Formation of Gubbio – la Bottacione shows many prominent 5 to 10 cm-thick cherty beds in the lower Campanian, overlain by a 5 m-thick marly interval (Fig. 4). Many stratigraphic studies, including pioneering magnetostratigraphy (Lowrie and Alvarez, 1977) and biostratigraphy based on foraminifera (Premoli Silva, 1977), have been published for the section. In this paper we use the stratigraphic data from Coccioni and Premoli Silva (2015) who recently revised the Upper Albian–Maastrichtian bio- and magnetostratigraphy of this Tethyan reference section.

The Furlo – Upper Road section (43°38'29"N 12°42'36"E) located north of the Umbria-Marche basin (Fig. 1) exposes pelagic carbonate deposits from the Jurassic to the Palaeocene (~300 m-thick). A well-defined magnetostratigraphy (Alvarez and Lowrie, 1984) and a U/Pb age from a bentonite layer identified within chron C33r (Mattias et al., 1988; Bernoulli et al., 2004), provide a stratigraphic framework for the Campanian–Maastrichtian interval. Slope deposits are expressed in the upper part of the section by the occurrence of more than 70 (10 to 100 cm-thick) white-coloured turbidites, and by a 12 m-thick slump at the base of chron C33n (Fig. 4).

#### 2.2.2. Tunisian Atlas: El Kef – El Djebil section

During the Campanian–Maastrichtian, the Saharan platform was located at ~18°N on the southern margin of the Tethys Ocean (Philip and Floquet, 2000; Fig. 1). The closure of the Tethys generated tectonic compressive and extensive domains; the Saharan platform belonged to an external domain of intracontinental deformation, far from the intra-oceanic deformation zone to the north (Aris et al., 1998; Boutib et al., 2000; Frizon de Lamotte et al., 2009; Bey et al., 2012).

The 500-m thick El Kef – El Djebil section, located in the Tunisian Atlas immediately to the north of the Saharan platform (36°10'37"N 08°44'05"E) corresponds to the Abiod Chalk Formation. The Campanian–Maastrichtian Abiod Formation comprises three members (Burollet, 1956; Jarvis et al., 2002; Fig. 3), from base to summit: white chalks with occasional calciturbidites (lower chalk 'bar' or Haraoua Member); bioturbated marls with common limestone beds (middle marl or Akhdar Member); and a yellow-greyish bioturbated chalky unit (upper chalk 'bar' or Ncham Member). The biostratigraphic framework is mainly based on well-preserved planktonic foraminifers, complemented by carbon isotope chemostratigraphy (Robaszynski et al., 2000; Jarvis et al., 2002; Mabrouk El Asmi, 2014).

## 3. Materials and Methods

### 3.1. Oxygen and carbon isotopes

New stable-isotope data were generated for the two Italian studied sections (Supplementary Data A, B). Wherever possible, samples were recovered for geochemical analyses every metre from the Gubbio – la Bottaccione section and every half metre from the Furlo – Upper Road section. Stable-isotope analyses of carbonate ($\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) were performed on bulk rocks collected along the whole of each section, from the Santonian–Campanian boundary to the Campanian–Maastrichtian boundary (Fig 4). Isotopic analyses were carried out at the Leibniz–Laboratory für Altersbestimmung und Isotopenforschung, Christian–Albrechts University, Kiel, Germany. Samples devoid of macrofossils were crushed in an agate mortar and pestle into fine and homogeneous calcite powders, which were reacted with 100% phosphoric acid at 70°C and analysed using a ThermoScientific MAT253 mass spectrometer, connected to a Kiel IV preparation device. Eleven samples from the Gubbio – la Bottaccione section were additionally analysed at the Biogéosciences Laboratory, University of Bourgogne Franche-Comté, Dijon, France. Here, calcite was reacted with 100% phosphoric acid at 90°C using a Multiprep online carbonate preparation line connected to an Isoprime mass spectrometer.

All isotopic values are reported in the standard δ-notation in per mil relative to V-PDB (Vienna Pee Dee Belemnite) by assigning a $\delta^{13}C$ value of +1.95‰ and a $\delta^{18}O$ value of −2.20‰ to NBS19. External reproducibility as determined by replicate analyses of laboratory standards was ±0.08‰ (2σ) for oxygen isotopes in both laboratories and ±0.05‰ (2σ) for carbon isotopes at Leibniz – Laboratory and ±0.04‰ at the Biogéosciences Laboratory.

## 3.2. Clay mineralogy

All bulk-rock samples were collected in the field and from the core storage facility with a regular sample spacing (Figs 2–4). Mineralogical analyses were performed at the Biogéosciences Laboratory, University of Bourgogne Franche-Comté. Clay minerals assemblages were identified by X-ray diffraction (XRD) on oriented mounts of non-calcareous clay-sized particles (<2 μm). The procedure described by Moore and Reynolds (2009) was used to prepare all samples to better compare the integrity of the dataset and to avoid discrepancies due to the process of quantification. Diffractograms were obtained using a Bruker D4 Endeavor diffractometer employing $CuK_\alpha$ radiation with a LynxEye detector and Ni filter, under 40 kV voltage and 25 mA intensity. For each sample, three preparations were analysed: after air-drying; after ethylene-glycol solvation; and after heating at 490°C for 2 hours. The goniometer was scanned from 2.5° to 28.5° 2θ for each run.

Clay minerals were identified by the positions of their main diffraction peaks on the three XRD runs (Table 1), while semi-quantitative estimates were produced in relation to their peak areas (Moore and Reynolds, 2009). Peak areas were determined on diffractograms of glycolated runs with MacDiff 4.2.5 software (Petschick, 2010). The percentages of kaolinite and chlorite were determined by deconvolution of the $d(002)_{kaolinite}$ and $d(004)_{chlorite}$ peak areas that appear respectively at 3.57 and 3.52Å and using the 7.1Å peak area common to both minerals. Beyond the evaluation of the absolute proportions of the clay minerals, the aim was to identify their relative fluctuations through the sections.

Measurement of the relative proportions of smectite and illite layers in the R0 mixed layers were performed on the diffractograms, following two methods: the procedure of Moore and Reynolds (2009) and the determination of the saddle Index after Inoue et al. (1989). In most cases, the procedure of Moore and Reynolds was performed on reflections 001/002 and 002/003. However, the presence of illite at 10Å stretches the 001/002 reflection and modifies the true position of 001/002. In this case, estimation of the smectite layers was determined only using the reflection 002/003.

### 3.3. Correlation of the studied sites: $\delta^{13}C_{carb}$ isotopic events

We have used global and local carbon-isotope events previously recognised in the Campanian to correlate sections and boreholes. During the Campanian, seven isotopic events have been identified.

- The Santonian–Campanian Boundary Event (SCBE), consisting of a global positive shift of $\delta^{13}C_{carb}$, with a varying amplitude of 0.3‰ to 2.9‰ (Jarvis et al., 2002; Gale et al., 2008; Wendler, 2013; Thibault et al., 2016b; Dubicka et al., 2017), has been widely recognised in successions throughout the Boreal, Tethyan, North Pacific and Central Atlantic realms (Table 2, Event 1). This event coincides with the C34/C33r chron boundary and the Highest Occurrence (HO) of the crinoid *Marsupites testudinarius*, both defining the Santonian–Campanian boundary (Italy, Gubbio – la Bottaccione, Premoli Silva and Sliter, 1994; Texas, Waxahachie Dam Spillway, Gale et al., 2008; Poland Bocieniec, Dubicka et al., 2017).

- The *papillosa* Zone Event (PZE) is a positive excursion of ~0.2‰ coincident with a medium-term $\delta^{13}C$ maximum, occurring in the mid-Lower Campanian *papillosa* zone at Lägerdorf and in the uppermost *Globotruncana elevata* zone (Chron 33r/33n boundary) on the Bottaccione section. Its stratigraphic significance still needs to be tested by additional high-resolution data sets (Thibault et al., 2016b; Sabatino et al., 2018; Table 2, Event 2).
- The Mid-Campanian Event (MCE), first described by Jarvis et al. (2002) on the $\delta^{13}C_{carb}$ curves of El Kef (Tethyan realm, Tunisia), Bidart (North Atlantic realm, France) and Trunch (Boreal realm, England, Jenkyns et al., 1994), is defined by a positive excursion of 0.3‰ occurring near the base of *Globotruncana ventricosa* planktonic foraminifera zone and the base of the upper Campanian (Table 2, Event 3). At Tercis-les-Bains, this event occurs at the base of chron C33n, comprising the Lowest Occurrence (LO) of *Rucinolithus magnus* and *Uniplanarius gothicus* nannofossils. This event has been recognised at a larger scale by Perdiou et al. (2016) in the North Sea.
- The Conica Event (CE; Perdiou et al., 2016) is a small negative excursion of $\delta^{13}C_{carb}$ of ~0.4‰, occurring at the base of the *conica-senior* macrofossil zone, in the Boreal and North Atlantic realms (Table 2, Event 4).

- The Late Campanian Event (LCE) is a global event, identified in the Tethyan, Boreal, Central Pacific, Indian Ocean and North Atlantic realms (Jarvis et al., 2002, 2006; Voigt et al., 2010, 2012; Thibault et al., 2012b; Sabatino et al., 2018). It consists of a marked negative excursion of $\delta^{13}C_{carb}$ with an amplitude varying between 0.3‰ and 1.3‰. It is located within the middle of chron C33n (Table 2, Event 5). This event occurs within (Gubbio, Voigt et al., 2012; Sabatino et al., 2018) and/or immediately above (El Kef, Jarvis et al., 2002; Tercis-les-Bains, Voigt et al., 2012, Chenot et al., 2016) the *Radotruncana calcarata* planktonic foraminifera zone (UC15 d-e nannofossil zone) in the Tethyan realm and in the mid-*Belemnitella mucronata* macrofossil zone in the Boreal realm. Perdiou et al. (2016) identified two steps in this isotopic event, called the pre-LCE and the main-LCE, which coincide with an increase of illite, kaolinite and chlorite in the Aquitaine basin (Chenot et al., 2016) and illite in the Paris basin (Deconinck et al., 2005).
- The Epsilon event (EE or C1-) was defined by Thibault et al. (2012a) as a negative excursion of ~0.25‰, followed by a positive shift of 0.2‰ and a second negative excursion of 0.1‰, which imparts to this event a sharp resemblance to the Greek letter ε. The event occurs slightly above the HO of *Eiffellithus eximius* in the Boreal, Tethyan and North Atlantic realms (Table 2, Event 6).
- The Campanian–Maastrichtian Boundary Event (CMBE, Voigt et al., 2010, 2012) is a global negative event with an amplitude ranging from 0.3‰ to 1‰, occurring in the reverse chron C32n2n, and identified in many sedimentary basins from the Tethyan to Pacific realms (Table 2, Event 7). This isotopic event was divided into three steps by Thibault et al. (2012a): CMBa (negative excursion of 0.6‰); CMBb (positive excursion of 0.2‰); CMBc (negative excursion of 0.4‰). The definition of the CMBE by Voigt et al. (2012) is different as it comprises the whole long-term decrease

in carbon isotopes ranging from the middle of chron C32n2n up to the lower half of chron C31r and is further described by 5 distinct small positive peaks that are superimposed on the long-term trend (CMBE1 to CMBE5).

### 3.4. Calcareous nannofossils biostratigraphy

The El Kef – El Djebil section constitutes one of the reference isotopic curves for the Campanian of the Tethys (Jarvis et al., 2002) but so far, no calcareous nannofossils biostratigraphy was available for that section. In this study, we analysed 17 samples from the archive of Jarvis et al. (2002) in order to establish a coarse calcareous nannofossils biostratigraphy that can be directly correlated to the already available isotope curve. Standard smear-slides were prepared following the methodology described in Bown (1998) and the biostratigraphy is based solely on presence/absence of individual taxa observed in crossed nicols at a magnification of x1000 on a Leica DM750P optical microscope. The CC nannofossil zonation of Sissingh (1977) modified by Perch-Nielsen (1985) and the $UC^{TP}$ (Tethyan Province) zonation of Burnett (1998) have been applied (Fig. 5).

### 3.5. Calcium carbonate content

Calcimetry was performed on the Gubbio – la Bottaccione and Cbr-7 samples at the Biogéosciences Laboratory, University of Bourgogne Franche-Comté, using a Bernard calcimeter. The samples were treated with hydrochloric acid and the $CO_2$ released was used to quantifying the percentage of $CaCO_3$. The data are reported on Fig. 3 and Supplementary Data C for the Cbr-7 borehole and in Supplementary Data B for the Gubbio – la Bottaccione section.

## 4. Results

### 4.1. Clay mineralogy

The clay fraction of the six studied sites is composed predominantly (often more than 80%) of R0 random illite/smectite mixed-layers; hereafter referred to as smectitic minerals (not represented on Figs. 2–4 to emphasize other clay minerals variations; Supplementary Data A, B; C, D, E, F). This result was expected, since Upper Cretaceous sediments are characterised by an abundance of these minerals, considered to be the background of the clay sedimentation (Deconinck and Chamley, 1995; Deconinck et al., 2005; Jeans, 2006; Chenot et al., 2016). Other clay minerals, occurring in significant proportions, include illite, chlorite, kaolinite and palygorskite.

#### 4.1.1. Stevns-2 and Adda-3 boreholes

Illite (less than 10% of the clay fraction) occurs in most samples from the Stevns-2 borehole. Traces of chlorite (less than 2%) are recorded between 349.9 and 251.7 m depth corresponding to beds of alternating chalk-marl and higher gamma-ray values of the upper Campanian; this interval is associated with a warm optimum preceding the early Maastrichtian cooling (Thibault et al., 2016a; Boussaha et al., 2017). Chlorite is absent above the Campanian–Maastrichtian boundary (Fig. 2; Supplementary Data D). The clay minerals assemblage of the Adda-3 borehole includes traces of illite and traces of kaolinite in all samples (Fig. 2; Supplementary Data E). Based on the methods of Moore and Reynolds (2009) and Inoue et al. (1989), estimation of the smectite layers in the IS R0 comprises between 50 and 80% in the Stevns-2 borehole (Supplementary Data D) and between 85 and 95% in the Adda-3 borehole (Supplementary Data E). The evolution of the percentage does not show any trend.

### 4.1.2. Cbr-7 borehole

According calcium carbonate content data, the percentage of the clay fraction is estimated to range between 5% from 75 to ~40 m depth to < 3% from ~40 m to the top of the core (Fig. 3; Supplementary Data C).

In the Trivières Chalk, at the base of the core, beside smectitic minerals, the clay assemblages consist of 20% illite, with small quantities of kaolinite that decrease from the base to the top of the formation (from less than 10% to traces). Kaolinite disappears in the overlying Obourg Chalk, while illite occurs in small proportions in this formation, together with traces of fibrous clays (palygorskite), clinoptilolite and talc. The Nouvelles Chalk is characterised by increasing proportions of illite with varying quantities of fibrous clays, clinoptilolite and talc (Fig. 3).

To estimate the percentage of smectite layers in the IS R0, the method of Moore and Reynolds (2009) could not be performed on the Cbr-7 borehole diffractograms because reflections 001/002 and 002/003 are poorly expressed. However, the method of Inoue et al. (1989) shows an evolution of the Saddle Index similar to the trend of illite. First, from 80 to 45 m depth, the saddle index displays a decreasing trend from 0.7 to 0.3, which means an increase of smectite layers in the IS R0 (from ~60 to ~80%). In a second part, the saddle index records an increasing trend between 45 – 10 m depth from 0.3 to 1, interrupted by highest values around 35 m, corresponding to smectite layers ranging from ~80 to ~50%. At the top of the borehole, from 7 to 3 m depth, the saddle index displays the lowest values around 0.5, equivalent to about 70% of smectite layers (Supplementary Data C).

### 4.1.3. Gubbio – la Bottaccione and Furlo – Upper Road sections

In the Gubbio – la Bottaccione section, from the base to 45 m, beside abundant smectitic minerals, the clay fraction consists of illite (10%) with occasional traces of kaolinite and chlorite (Fig. 4; Supplementary Data B), while from 50 to 70 m, kaolinite and chlorite occur systematically and increase up to maxima of >5% along with abundant illite (50%). In the uppermost part of the section, the proportions of kaolinite and chlorite decrease and kaolinite essentially disappears from 80 m upwards. From the base to 50 m, the percentage of smectite layers in the IS R0 is estimated to ~70%, whereas from 50 to 80 m, it decreases until ~50%. From 80 m to the top of the section, smectite layers increase again to ~65 % (Supplementary Data B).

From the base of the Furlo section to 44 m, the percentage of illite is around 10% with traces of kaolinite (Fig. 4; Supplementary Data A). From 57 m, kaolinite increases significantly, rising upwards to >10% at 75 m, together with more abundant illite and traces of chlorite. Interestingly, the onset of this major mineralogical change coincides with the appearance of turbidites above a 12 m-thick slump. The percentage of smectite layers in the IS R0 follows an opposite trend compared to illite until 80 m: from the base of the section to 44 m the highest percentage of smectite layers is estimated to ~70% and then progressively decreases down to ~50%. However, from 80 m to the top of the section, the percentage of smectite layers in IS R0 seems to increase again up to ~70% (Supplementary Data A).

### 4.1.4. El Kef – El Djebil section

In the sediments from El Kef – El Djebil section, the evolution of the clay fraction, again dominated by smectitic minerals, is divided into several mineralogical zones (Fig. 3; Supplementary Data F). Traces of illite are recorded in most samples. By contrast, kaolinite and chlorite are more abundant in 3 intervals. In the first interval, from 0–60 m, the percentages of kaolinite and chlorite diminish and these minerals disappear upward. In the second interval, from 80–330 m, the proportion of kaolinite increases to a maximum of 10% around 180 m, and then decreases progressively to 330 m. Chlorite shows a similar trend, with maximum values of around 5% at 180 m. Interestingly, the highest percentages of chlorite and kaolinite occur at the transition between the lower white chalks and the bioturbated marls. In the third interval, from 410–430 m, the proportion of kaolinite again rises up to 10%, along with traces of chlorite, and then falls from 430–465 m. However, the values in the third interval must be interpreted with caution, because of the high proportion of Si which distorts the peak area on the diffractogram required for percentage calculation (Si/Al ratio determined by an ICP-AES by Mabrouk El Asmi, 2014; Fig. 3).

## 4.2. Isotope analyses

### 4.2.1. Gubbio – la Bottaccione section

Bulk-rock $\delta^{13}C$ values range from about 2.1 to 2.7‰ through the Gubbio – la Bottaccione section (Fig. 4; Supplementary Data B). Three moderate isotopic excursions may be identified. From 1.2–9.5 m, a 0.3‰ positive shift is observed at the Santonian–Campanian transition and the C34/C33r chron boundary. A second positive shift of 0.4‰ starts at 23.8 m and ends at 40.3 m, before the first exposure gap, at the base of the *Contusotruncana* *plummerae* zone. After increasing from 2.3 to 2.6‰ up to 61 m, $\delta^{13}C$ values decrease down to about 2.4‰ up to 83 m. This decrease mostly occurs in the *Radotruncana calcarata* zone and the upper part of chron C33n. From 95.8 m to the top of the section, the bulk-rock $\delta^{13}C$ values display a decreasing trend of 0.2‰, which coincides with the Campanian–Maastrichtian transition, within the *Gansserina gansseri* zone.

Bulk-rock $\delta^{18}O$ values display an increasing trend from values of about -3.0‰ at the base of the Gubbio – la Bottaccione section, to values of about -2.2‰ around 40 m, and do not show any further trend for the remaining of the section (Fig. 4; Supplementary Data B).

#### 4.2.2. Furlo – Upper Road section

Bulk-rock $\delta^{13}C$ values range from about 2.0 to 2.9‰ in the Furlo – Upper Road section (Fig. 4; Supplementary Data A). Between 1.3 and 5.9 m, a first two-step positive excursion of 0.4‰ coincides with the C34/C33r chron boundary and the Santonian–Campanian transition. This excursion is followed by a negative excursion of about 0.4‰ with $\delta^{13}C$ values reaching 2.2‰ at 17 m. An increase in $\delta^{13}C$ values is then recorded, with maximum values of about 2.9‰ at about 30 m. From about 30 m to 44 m, the data show an overall decreasing trend from values of about 2.9‰ to values of about 2.6‰, with one sample at the top of the interval displaying a value of 2.4‰. It can be noted that this interval coincides with the first occurrence of turbidites.

Above the 12 m-thick slump in the middle part of the studied section, bulk-rock $\delta^{13}C$ data decrease from values of 2.6‰ at 56.7 m to values on average 2.3‰ at 92 m. This trend is interrupted by a two-step negative excursion of 0.5‰, from 57.7 to 65.5 m, and from 65.5 to 72.5 m, occurring in the middle of chron C33n. Above 92 m, following a small positive excursion of about 0.2‰, $\delta^{13}C$ values decrease again upwards, from $\delta^{13}C$ values of 2.5‰ at

93 m, and down to minimum values of 2‰ at the top of the section. The onset of this decrease coincides with the C33n1r/C32n2n chron boundary.

The $\delta^{18}O$ values remain quite stable in the lower part of the section, from the base to 44 m, with values around -2‰ on average (Fig. 4; Supplementary Data A). In the upper part of the section, they display a slight increase, from values of about -2‰ to values of about -1.5‰ around 80 m, before decreasing again to about -2‰ at the top of the section.

### 4.3. Nannofossils bioevents and biozonation of El Kef

A total of 17 samples of the El Kef section were studied here. Sample 81 (98.55 m) was barren, but the remainder of the samples yielded numerous late Cretaceous specimens. In total 61 individual calcareous nannofossil species were recognized in this study, a number which is rather low for the Campanian as this stage is characterised by the highest species richness of the whole Mesozoic (Bown, 2004). The low diversity recorded in El Kef section is likely the result of the preservation of the assemblage that is at best moderate. *Watznaueria barnesiae*, *Cribrosphaerella ehrenbergii*, *Prediscosphaera cretacea* and *Zeugrhabdotus bicrescenticus* are common.

The studied samples belong to the interval from zone CC18-CC19 to CC24 of Sissingh (1977) and UC14a$^{TP}$-UC15b$^{TP}$ to UC18 of Burnett (1998) due to the presence of *Broinsonia parca constricta* in the first sample (K1, 2.2 m) and the HO of *Tranolithus orionatus* in sample K447 (474.6 m). According to this applied biozonation, the age of the studied interval should range from the late early Campanian to early Maastrichtian. However, the inconsistency in the order of last occurrences recorded in the upper part of the section with respect to global schemes as well as with respect to other stratigraphic considerations, suggests that the zonations are hardly applicable to the section and that the whole studied succession remains restricted to the Campanian only (see Discussion). A summary of the main results is provided in Fig. 5 (range chart with zonation and bioevents).

## 5. Discussion

### 5.1. Influence of diagenesis

A prerequisite for the use of clay minerals for palaeoenvironmental reconstructions is that they should mainly have a detrital origin. Smectitic minerals, which are the background of clay sedimentation in most latest Cretaceous open marine environments, are very sensitive to burial diagenesis. Illitization processes start when the temperature reaches about 60°C, (Kübler and Jaboyedoff, 2000; Kübler and Goy-Eggenberger, 2001) and IS R0 are progressively transformed into I/S R1, then R3, and finally into illite (Środoń, 2009). In each studied section, the illitization process is considered as subsidiary because of the high abundance of IS R0 throughout the successions (Supplementary Data A, B, C, D, E, F; Delissanti et al., 2010). In addition, low $T_{max}$ values, comprised between 402° and 433°C, have been determined in previous studies for the "Bonarelli level" (OAE 2) of the Furlo section (Scaglia Rossa Formation), that corresponds to the immature zone of the organic matter, thereby suggesting negligible burial diagenesis consistent with persistence of IS R0 (Mort et al., 2007; Delissanti et al., 2010).

The $\delta^{13}C$ values of sediments from the Gubbio – la Bottaccione and Furlo – Upper Road sections comprised between 2.0 and 3.0‰, match $\delta^{13}C$ values typically observed in latest Cretaceous marine sediments of the Boreal and Tethyan realms (Jenkyns et al., 1994; Jarvis et al., 2002; Voigt et al., 2012; Figs. 6, 7). A cross-plot between $\delta^{13}C$ and $\delta^{18}O$ values shows close agreement for carbon between Furlo and Gubbio but a difference in oxygen, with samples from Gubbio presenting lower values. A Spearman's coefficient was computed for each dataset to test the existence of a correlation within the data of each section. This method was chosen because of the non-linear nature of the relationship between the two variables, $\delta^{13}C$ and $\delta^{18}O$ (Chenot et al., 2016). A value of 1 indicates a perfect correlation, a value of -1 a perfect anti-correlation, while 0 indicates an absence of correlation. The two datasets generate Spearman's coefficients of $r_s$ = -0.66 for Furlo and $r_s$ = +0.11 for Gubbio (Fig. 6), pointing to an absence of a significant correlation between $\delta^{13}C$ and $\delta^{18}O$ values within each dataset.

The isotope data do not exhibit any inverted J curve in $\delta^{13}C$–$\delta^{18}O$ space that could reflect diagenesis involving fluid-rock interactions in addition to physical mixture of different diagenetic mineral phases (e. g. Bishop et al., 2014). This supports an absence of extensive diagenesis affecting both isotopic systems, and argues in favour of a preservation of $\delta^{13}C$ values. By contrast, the markedly lower $\delta^{18}O$ values recorded at Gubbio, which presents a deeper depositional environment compared to Furlo, likely indicate an impact of diagenesis on oxygen isotopes, although probably limited considering that the values fall within the range of shallow buried pelagic carbonate successions of comparable age elsewhere (e.g. Voigt et al., 2010; Fig. 7).

### 5.2. Identification and redefinition of carbon-isotopic events

In order to correlate the studied sections and boreholes, we used seven carbon-isotopic events, namely the: (1) Santonian–Campanian Boundary Event; (2) *papillosa* Zone Event; (3) Mid-Campanian Event; (4) Conica Event; (5) Late Campanian Event; (6) Epsilon Event and (7) Campanian–Maastrichtian Boundary Event, described in Section 3.3 (Fig. 8; Table 2).

### 5.2.1. Identifications of carbon-isotopic events in the Gubbio – la Bottaccione and Furlo – Upper Road sections

A 0.4‰ positive shift in the basal Gubbio – la Bottaccione section (between 1.2 and 9.5 m) and a two-step positive carbon-isotopic excursion of 0.4‰, recorded on the Furlo – Upper Road section (between 1.3 and 5.9 m), coincide with the Santonian–Campanian transition and the C34/C33r chron boundary respectively and are therefore attributed to the SCBE (Figs. 4; 8).

A sharp increase of 0.4‰ in the $\delta^{13}C$ curve identified within the mid-Lower Campanian in the Gubbio – la Bottaccione section, starting at the base of the chron C33n and ending at the base of the *Contusotruncana plummerae* zone, is assigned to the *papillosa* Zone Event (cf. Thibault et al., 2016b; Sabatino et al., 2018). This excursion is not visible in the Furlo – Upper Road section profile where it may be obscured or missing due to the slump interval (Figs. 4, 8).

The MCE has been previously well-defined on the Bottaccione section, occurring above the LOs of *Rucinolithus magnus* and *Uniplanarius gothicus*, but is not recorded on the isotopic curve of this study, maybe because of the low amplitude of this isotopic event, around 0.2‰ (Thibault et al., 2016b; Sabatino et al., 2018).

The LCE is well defined in the Gubbio – la Contessa section (Voigt et al., 2012) and in the Gubbio – la Bottaccione section (Sabatino et al., 2018; Figs. 4, 8) with an amplitude of around 0.4‰. However, in this study, the LCE seems to be poorly expressed on our Gubbio – la Bottacione profile, likely because of the occurrence of several gaps in the sedimentary record of this section. However, the decreasing trend of 0.25‰ from 61 to 83 m identified in the la Bottaccione section in the upper part of the chron C33n, including the *Radotruncana calcarata* zone (Figs. 4, 8), is tentatively associated to the target horizon where the LCE could be expected. In the Furlo – Upper Road section, the two-step negative excursion of 0.5‰, above the 12 m-thick slump and occurring in chron C33n may be ascribed to the pre-LCE and main-LCE (Figs. 4, 8).

The onset of the CMBE is well represented in both the Gubbio – la Bottaccione (starting at 98.5 m) and Furlo – Upper Road (starting at 94.1 m) sections (Figs. 4, 8).

### 5.2.2. Identification and attribution of carbon-isotope events from other sections

#### 5.2.2.1. El Kef – El Djebil section

The calcareous nannofossil record of the upper part of the El Kef section (335 to 496 m) is difficult to interpret. Voigt et al. (2012, Fig. 6) documented a consistent succession of closely spaced nannofossil HOs that occur in the Lower Maastrichtian at Gubbio and Lägerdorf-Kronsmoor-Hemmoor, all within the upper part (their intervals 3 – 5) of the CMBE: *U. trifidus*, *B. parca constricta*, *T. orionatus*. The consecutive datum levels lie on a marked falling trend in the $\delta^{13}C$ curve, which precedes a sharp rise in values towards the Mid-Maastrichtian Event (MME), above. The same succession of nannofossil disappearances at El Kef are spread through >100 m of the Ncham Chalk, largely within an interval of relatively high $\delta^{13}C$ values (Figs. 3, 8) above the negative excursion defining the LCE.

The HO of *U. trifidus* has been recently considered as the best nannofossil marker for the Campanian/Maastrichtian boundary and was used to subdivide UC zone 16 into subzones UC16a$^{TP}$ and UC16b$^{TP}$ (Thibault, 2016). The HO of *U. trifidus* at 403 m would thus suggest that this boundary lies close to that stratigraphic height, and that the negative excursion recorded between 339 and 390 m corresponds to the CMBE, not the LCE. However, the HO of *B. parca constricta*, which marks the top of UC18 and should thus be recorded higher up at

El Kef was found here below the HO of *U. trifidus*. Moreover, a narrow range of the curved spine nannolith at El Kef, straddling the negative excursion, contradicts other nannofossil results and the identification of this excursion as the CMBE. Indeed, the HO of the curved spine is generally restricted to the late Campanian, and correlates to the top of the LCE (Thibault et al., 2012a; Thibault, 2016; M.R. Razmjooei and N. Thibault, pers. comm., unpublished results from Iran). It is therefore possible that the HOs of *B. parca constricta* and *U. trifidus*, which are nearly identical between El Kef (this study) and Kalaat Senan (Robaszynski et al., 2000) are recorded in the region of the Kasserine Island much earlier than in the rest of the Tethys due to local environmental conditions.

The negative $\delta^{13}C$ excursion between 339 and 390 m at El Kef lies in the upper part of the Akhdar Marl Member *sensu* Jarvis et al. (2002). Robasyzynski et al. (2000) recorded the Upper Campanian ammonite *Nostoceras (Nostoceras) hyatti* above this in the lowest part of the overlying Ncham Chalk Member. The index ammonite taxon *Nostoceras (Bostrychoceras) polyplocum* ranges through the lower part of the Akhdar Marl, overlapping with the base of the *G. calcarata* foraminifera Zone, above. The negative $\delta^{13}C$ excursion occurs above the top of the *G. calcarata* Zone and below the base *N. hyatti*. This stratigraphic relationship between the excursion and the macrofossil datum levels is essentially identical to that recorded for the Late Campanian Event (LCE) in the Tercis-les-Bains GSSP by Voigt et al. (2012), and is well below the base of the CMBE. These data support the original interpretation of Jarvis et al. (2002) that the negative $\delta^{13}C$ excursion at El Kef is the LCE not the CMBE.

The planktonic foraminifera biostratigraphy is not definitive: *G. gansseri* occurs very infrequently at its lowest occurrence at Kalaat Senan (Robaszynski et al. 2000, Figs. 3, 8), and is only recorded consistently in the boundary interval between the Akhdar and Ncham Members ('Gorbeuj Member' of Robaszynski et al. 2000). The first consistent occurrence correlates to above the negative excursion at El Kef (Jarvis et al. 2002, Fig. 2). Unfortunately, *G. gansseri* was not identified in the low-resolution foraminifera study of El Kef (El Djebil) by De Cabrera (in Jarvis et al. 2002).

Finally, attributing the main negative excursion at El Kef to the CMBE rather than the LCE would necessitate major changes in sedimentation rate within the section that are not supported by coincident lithological variation, and/or challenges the stratigraphic completeness of the logged succession. In conclusion, we retain here the original carbon isotope event assignments of Jarvis et al. (2002), while acknowledging the challenges of the stratigraphy that warrant further study.

#### 5.2.2.2. Cbr-7 borehole

The short 1‰ negative excursion of $\delta^{13}C$ measured by Richard et al. (2005) in the Cbr-7 borehole (Figs. 3, 8) was not ascribed to any recognized isotopic event. The Mn peak observed in this borehole corresponds to a glauconite-bearing hardground, which points to the existence of a condensed interval and/or hiatus, consistent with Mn accumulation. In this case, the 1‰ excursion recorded in $\delta^{13}C$ values may correspond to the LCE, according to its occurrence within the lower part of the upper Campanian.

However, the hardground at the summit of the Trivières Chalk occurs in the basal *Belemnitella mucronata* zone (Robaszynski et al., 2001; Richard et al., 2005), i.e. very close to the base of the Upper Campanian as calibrated in NW Europe. Interestingly, a succession of hardgrounds occurs at a very similar stratigraphic level in the Trunch borehole of Eastern England, called the Trunch Hardgrounds by Jarvis et al. (2002). The MCE is placed immediately above this condensed interval in the Trunch borehole. If these hardgrounds are equivalent to the Trivières hardground, then the negative $\delta^{13}C$ excursion in the Cbr-7 borehole correlates to the negative excursion immediately below the Trunch Hardgrounds, and the small peak above the negative excursion in Cbr-7 would be the MCE. Decreasing-upward values at the top of Cbr-7 in the Nouvelles Chalk can then be interpreted to represent the lower part of the LCE, which we used in this study for the stratigraphic correlation (Figs. 3, 8).

## 5.3. Clay minerals origin in the Tethyan realm

The origin of smectitic minerals in oceanic sediments is controversial and has been debated by Chamley (1989) and Thiry (2000). The abundance of these minerals in Cretaceous sediments, especially from cores drilled in the Atlantic Ocean, is interpreted differently by the authors. Smectitic minerals are considered to be detrital in some studies, based on their chemical composition (Al-Fe beidellite), which is similar to smectitic minerals formed in soils, on their REE element profiles, and on their strontium isotope composition (see Chamley, 1989; Chamley et al., 1990). Alternatively, a volcanogenic or early diagenetic origin of smectitic minerals has been proposed based on the common occurrence of the paragenesis smectite opal-CT-clinoptilolite (Pomerol and Aubry, 1977; Christidis, 1995; Madsen and Stemmerik, 2010). The controversy also arises from the coeval occurrence of smectite-poor continental successions and smectite-rich marine sediments. This paradox has been tentatively explained by the massive neoformation of smectitic minerals in oceanic basins or by the transformation of detrital clay particles into smectitic minerals (Thiry and Jacquin, 1993). Other authors have suggested that differential settling processes are responsible for the smectitic minerals enrichment in (hemi-) pelagic environments (Chamley, 1989).

In the clay fraction of the Upper Cretaceous Chalk, it is now clear that the smectitic minerals correspond to a mixture of detrital I/S, authigenic lathed smectitic minerals preferentially formed in slowly deposited sediments, and smectitic minerals deriving from the submarine weathering of volcanic glass shards (Deconinck and Chamley, 1995; Jeans, 2006). Environmental conditions during the Late Cretaceous favoured dominantly smectitic sedimentation: high sea level; low topographic relief on continental areas; relatively seasonally contrasted humid climate; low sedimentation rates; and recurrent volcanism expressed by the common occurrence of bentonite layers (Chamley et al., 1990; Deconinck and Chamley, 1995).

Chlorite and illite are generally directly reworked from igneous or metamorphic continental basement rocks and are therefore commonly considered to be primary minerals (Chamley, 1989; Weaver, 1989; Ruffell et al., 2002). Consequently, their proportions increase either during tectonic rejuvenation, sea-level fall, or under dry climate when chemical weathering is reduced. By contrast, the pedogenic formation of kaolinite occurs under warm and humid conditions suitable for high rates of chemical weathering (Ruffell et al., 2002). However, this mineral may be also reworked together with primary minerals (e.g. illite, chlorite) from ancient kaolinite-bearing sedimentary rocks (Deconinck and Vanderaveroet, 1995). In that case, kaolinite cannot be used as evidence for the existence of humid climate. Coupling or decoupling in the variation of primary minerals and of kaolinite, however, may help differentiate between pedogenic vs. reworked origins, thereby indicating the reliability of kaolinite as a palaeoclimate indicator.

An important feature that is highlighted by clay mineral assemblages of the studied sections is the absence of significant amounts of kaolinite in the northernmost sections located in the Danish basin and the North Sea (Fig. 9). By contrast, kaolinite occurs in the three sections located south of 30°N (El Kef – El Djebil and Umbria-Marche localities). The two sections located latitudinally in-between, in the Paris and Mons basins, present intermediate characteristics with kaolinite present in the Cbr-7 borehole, but absent in the Poigny borehole. This broad latitudinal zonation may result from a climatic control, as humid/arid conditions are known to impact clay mineral assemblages (Ruffell et al., 2002; Dera et al., 2009). The common occurrence of bauxite reported in the general area of Gubbio, Furlo, and Tercis-les-Bains (Bárdossy and Dercourt, 1990), supports at least a partial pedogenic origin of kaolinite observed in these sections (Fig. 10).

Based on the distribution of kaolinite and bauxite along the palaeolatitudinal transect, climatic zones are proposed in the Tethyan/Boreal realms during the Campanian (Figs, 10, 11). Between 20° and 35°N, the clay fractions are characterised by the occurrence of kaolinite and iron (oxyhydro)oxides (e.g. hematite in the Scaglia-Rossa Formation; Channell et al., 1982), corresponding to intense weathering under warm and humid climate conditions. These climatic conditions are also highlighted by the distribution of Late Cretaceous bauxites which are abundant in southern France and in the Dinarides at palaeolatitudes below 35°N (Charvet, 1978; Bárdossy and Dercourt, 1990; D'Argenio and Midszenty, 1995; Fig. 10). North of 35°N, clay assemblages are mainly dominated by smectitic minerals and illite (with traces of chlorite and kaolinite), and the absence of bauxite indicates limited chemical weathering, consistent with seasonally contrasted climate.

However, variations in primary mineral proportions (chlorite and illite) mirror those of kaolinite, pointing to the additional presence of reworked kaolinite. A similar co-variation of primary minerals and kaolinite is identified at El Kef, again suggesting the predominance of kaolinite reworked from old rocks that cropped out on nearby emergent landmasses. To our knowledge, no major bauxite deposits are reported for the Upper Cretaceous in that area (Tardy et al., 1991; Monsels, 2016), which was located in an arid zone according to the distribution of vegetation (Otto-Bliesner and Upchurch, 1997; Chumakov, 2004). These observations suggest that kaolinite is dominantly reworked together with primary minerals in all sections south of 30°N. Consequently, clay sedimentation was not solely controlled by climate but involved an additional factor, which could be linked to the tectonic instability of the southern margin of the Tethys, or to the long-term trend of sea-level fall that characterises the Late Cretaceous.

### 5.4. Diachronous fluctuations of detrital clay minerals with palaeolatitude

A striking feature arising from the overall clay mineralogical results of all studied sections is the earlier occurrence of kaolinite on southern Tethyan margin sites compared to the northern domain (Fig. 9). At El Kef, three intervals of increased detrital minerals (chlorite and kaolinite) are recorded: from the transition between the lower to middle Campanian (within CC19 and the lower part of CC20 nannofossil biozones); from the base of CC21 to the top of the *Radotruncana calcarata* foraminifera zone; and from the base to the top of CC23 (Fig. 3). In the Umbria-Marche basin, the onset of detrital input of clay minerals occurs in youngest sediments dated to the upper part of the middle Campanian (Fig. 4). At Tercis-les-Bains, illite and kaolinite contents increase at the base of the pre-LCE, while a coeval increase of illite is recorded in the Paris Basin (Poigny borehole; Chenot et al., 2016, Fig. 9). Consequently, the studied sections point to a general increase in detrital input during the Campanian that is diachronous, with an earlier onset in the southern sections (El Kef, Furlo, Gubbio) than those immediately to the north (Tercis-les-Bains, Poigny).

In the northernmost sections (Poigny, Adda-3 and Stevns-2), no clear trend is apparent in the abundance of detrital clay minerals, except for Cbr-7 (Fig. 9). At that site, an increase in detrital input is recorded by an increase in illite and talc proportions in the upper part of the borehole. This contrasts with the other sites where the increase in detrital inputs is marked by increasing kaolinite proportions. This difference may result from a different climatic context that is evidenced by the transition from kaolinite-bearing clay fraction at the base of the borehole toward sediments containing palygorskite, suggesting an evolution from relatively humid to more semi-arid conditions. It is however worth noting that a hardground separates the two intervals in Cbr-7, pointing to the existence of a significant hiatus, the duration of which cannot be evaluated because of the lack of biostratigraphic markers. The large uncertainties on the stratigraphic framework preclude further temporal comparison of the depicted increase in detrital inputs with that recorded at the other studied sites (Fig. 9).

### 5.5. A tectonic versus climatic control of clay sedimentation

Although the long-term sea-level fall recorded during the Late Cretaceous could have contributed to the general increase in detrital inputs depicted here (Haq et al., 1988; Hardenbold et al., 1998; Haq, 2014), their diachronism between sites strongly suggests the existence of additional processes at play.

The Late Cretaceous was characterised by compressive events around the Tethys, linked to the northward motion of Africa toward Eurasia (Kley and Voigt, 2008; Frizon de Lamotte et al., 2011; Jolivet et al., 2016). During the Campanian – Maastrichtian interval, large areas of emerged land and newly created relief in central Europe and in the western Tethyan realm (e.g. southern Carpathians, east-Pyreneans, inverted Mid-Polish Anticline, High-Karst) delivered detrital material to the adjacent sedimentary basins (Charvet, 1978; Willingshofer et al., 2001; Kley and Voigt, 2008; Voigt et al., 2008; Melinte-Dobrinescu and Bojar, 2010; Oms et al., 2016; Figs. 1, 10, 12).

On the southern Tethyan margin, evidence of tectonic instability as early as the earliest Campanian, is suggested by the occurrence of synsedimentary faults, slumps and slope instability features in sedimentary successions from northeastern Tunisia (Boutib et al., 2000; Bey et al., 2012). This tectonically driven sedimentation persisted throughout the Campanian and possibly into the Maastrichtian, as illustrated by syndepositional faulting and gravity flow deposits in the Abiod Chalk (Bouaziz et al., 2002; Dlala 2002; Negra, 2016). In the southernmost section of El Kef, the detrital influence occurring in the middle Campanian might be linked to a tectonic rejuvenation of nearby continental areas (e.g. Kasserine Island, Kadri et al., 2015; Figs. 1, 10, 12).

At El Kef, several kaolinite- and chlorite-enriched detrital intervals are recognised, separated by intervals devoid of typical detrital minerals. This may reflect distinct tectonic pulses affecting this segment of the southern margin (Figs. 3, 12). The kaolinite-rich interval within the *G. ventricosa* zone at El Kef is coeval with the occurrence of rudist-bearing olistolith beds on the NE margin of Kasserine Island (Negra et al., 2016), further highlighting a phase of platform destabilisation.

In central Italy (Umbria-Marche basin), detrital input of kaolinite and chlorite started later, in the earliest part of chron C33n (Figs. 4, 12). Volcanic activity in the area is highlighted by the preservation of a bentonite layer in the *G. elevata* zone (more precisely in the lower Campanian CC18 nannofossil biozone; Mattias et al., 1988; Fig. 4). The ashfall originated from an active volcanic centre related to a subduction zone located to the east of the Umbria-Marche basin, in the Dinarides (Charvet, 1978), and points to active tectonism in this region (Bernoulli et al., 2004; Schmid et al., 2008). In the Furlo – Upper Road section, the occurrence of the bentonite layer is followed by slope destabilisation features including decimetric turbidite beds and a 12 m-thick slump, suggesting long-lasting tectonic instability (Fig. 4).

Within the same tectonic and climatic context, additional differences in clay mineralogy between geographically close sections are evidenced in the Umbria-Marche basin. Indeed, a comparison between the Gubbio – la Bottaccione and Furlo – Upper Road sections shows that kaolinite is more abundant at Furlo, which was located on a slope at shallower depth than Gubbio (Fig. 4). This difference is emphasised by the occurrence of numerous slope deposits including slumps and turbidites at Furlo, while basinal deposits characterised the Gubbio section. This clay mineral distribution is attributed to differential settling of kaolinite which likely originated from the east, possibly from the High Karst (Gibbs, 1977; Charvet, 1978; Figs. 1, 12).

A local tectonic influence coinciding with the LCE is also recorded in the Tercis-les-Bains section where chlorite, illite and kaolinite increase (Figs. 9–12). This detrital event is likely due to a tectonic pulse linked to the Pyrenean compressional phase between Iberia and southern Europe (Laurent et al., 2001; Vergés et al., 2002; Oms et al., 2016). A coeval event characterised by increasing proportions of illite is recorded in the Paris Basin (Poigny borehole, Deconinck et al., 2005; Figs. 9–12), potentially related to a compressive event (Mortimore and Pomerol, 1997). Slump deposits recorded in the Anglo-Paris Basin in the middle Campanian (Gale et al., 2015) could also relate to local tectonic instability.

By contrast, in the northern part of the studied transect, there are no clear detrital events similar to those recorded in the southern sections, despite the presence of active tectonism in this region affected by inversions at the time (Kley and Voigt, 2008; Voigt et al., 2008), probably because this area was too far away from emerged areas (Figs. 9–12). Thus, the climatic control on clay sedimentation is more clearly expressed at these sites. In the Mons basin, during the late Campanian, the progressive decreasing proportion of kaolinite was followed by the occurrence of palygorskite, which reflects the establishment of increasingly semi-arid climatic conditions (Figs. 3, 9–12). However, traces of talc present in the late Campanian could be related either to a change of detrital sources or to the generation of newly exposed areas by the extensive coeval tectonism recorded in the basin (Vandycke and

Bergerat, 1989), which is consistent with the coevally increase of illite in this basin. In the Danish North Sea, similar semi-arid to semi-humid climatic conditions are consistent with the abundance of IS R0, which constitutes the entire clay fraction of uppermost Campanian sediments (Figs. 2, 9–12).

5.6. Carbon cycle and continental weathering

Carbon cycle changes and continental weathering had been tentatively linked through the observed correspondence between the carbon isotope excursion defining the LCE and enhanced terrigenous inputs, identified by a coeval increase in kaolinite, chlorite and illite proportions at Tercis-les-Bains and Poigny (Chenot et al., 2016). The new results presented here, which include data from sections located over a wider range of palaeolatitude, show that this relationship does not hold. Indeed, the diachronous nature of detrital supplies evidenced here, interpreted as reflecting the northward progression of tectonic deformation, results in a decoupling between the carbon-isotope excursions and the evolving clay minerals assemblages.

However, our new clay mineralogical data highlight enhanced continental weathering throughout the whole Campanian stage, with a diachronous onset from the south to the north (Figs. 10, 12). As silicate weathering is known to promote atmospheric $CO_2$ drawdown (Berner, 1990, 2004; Berner and Kothavala, 2001), our new data hint to a potentially major role of incipient orogenic processes on Late Cretaceous long-term cooling. The data compiled by Royer et al. (2012) and Franks et al. (2015) highlight lower $pCO_2$ levels in the Campanian and Maastrichtian than during the Albian to Turonian interval, but the temporal resolution is not sufficient to discuss correlations between $pCO_2$ fluctuations, tectonic phases and the evolution of clay minerals assemblages depicted in our study. Decreasing atmospheric $CO_2$

levels during the Late Cretaceous have been repeatedly associated to reduced outgassing $CO_2$ flux from mid-ocean ridge volcanism and arc magmatism (Berner et al., 1983; Jones et al., 1994; Godderis and François, 1995; Berner, 2004; McKenzie et al., 2016). Without excluding the role of reduced $CO_2$ volcanic outgassing, our new results highlight an additional important mechanism that may have contributed to the Late Cretaceous cooling. Although initiation of the Tethyan closure began during the mid-Cretaceous, the Santonian–Campanian is marked by a change of direction and faster motion of African toward Eurasia (Bosworth et al., 1999; Guiraud and Bosworth, 1999; Frizon de Lamotte et al., 2011; Jolivet et al., 2016). The Campanian is also characterised by an acceleration of the long-term climatic cooling recorded during the Late Cretaceous (Cramer et al., 2009; Friedrich et al., 2012; Linnert et al., 2014), which coincides with enhanced detrital inputs depicted by clay minerals in our study. This temporal coincidence between an acceleration of cooling, tectonic pulses, and clay detrital input evolution further argues for a significant impact on climate of plate tectonics linked to Africa-Eurasia convergence. Although the importance of this process remains to be tested quantitatively using geochemical models, our work opens new perspectives on the understanding of the Late Cretaceous climate cooling.

## 6. Conclusion

New results on clay minerals assemblages of Campanian sediments from six sections and boreholes ranging from the southern Tethyan margin to the Boreal realm, provide the first insights on the evolution of continental weathering at the Tethyan scale during a long underexplored time interval (11 Myr).

We propose a climatic zonation in the west Tethyan to Boreal realms during the late Campanian: (1) a semi-humid climate belt north of 35°N, based on the sporadic occurrence of kaolinite, the occurrence of palygorskite and the high percentage of smectitic minerals; (2) a warm and humid climate belt, based on the occurrence of kaolinite and bauxite between 35°–20°N; and maybe (3) a semi-arid zone south of 20°N, adjacent to the Saharan platform, based on the abundance of smectitic minerals, with kaolinite interpreted here as being reworked from the basement.

Superimposed on this latitudinal climate distribution, we have identified detrital events in several basins. These events resulted from weathering of emerged continental areas, that we relate to the main tectonic active zones, and more specifically to the large subduction zone of the central Tethyan realm between African and Eurasian plate (Umbria-Marche basin, Furlo and Gubbio sections), the extensional basins in southern Tethys (Saharan platform margin, El Kef section), and the collision between the Iberian and the Eurasian plates (Aquitaine basin, Tercis-les-Bains section). These tectonic instabilities, associated with a warm and humid climate, likely led to enhanced chemical weathering of the newly created continental relief.

The northward migration of an enhanced detrital flux evidenced by our new records, linked to the progression of compressional deformation, reflects the closure of the Tethys due to the northward motion of Africa. As chemical weathering of silicate induces $CO_2$ consumption, we suggest that Late Cretaceous cooling was partly linked to enhance continental weathering.

## 7. Acknowledgments

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## FIGURES

**Figure 1:** Palaeogeography of the Campanian–Maastrichtian of the west Tethyan and south Boreal realms (modified from Philip and Floquet, 2000). Location of the sites studied: [A] Adda-3 borehole; [C] Cbr-7 borehole; [F] Furlo – Upper Road section; [G] Gubbio – la Bottacione section; [K] El Kef – El Djebil section; [P] Poigny borehole; [S] Stevns-2 borehole; [T] Tercis-les-Bains section. The green squares represent previously published data (Deconinck et al., 2005; Chenot et al., 2016) used in this study.

**Figure 2:** Stratigraphy and clay mineralogy of the Campanian–Maastrichtian in northern boreholes. (A) Clay mineralogical data (this study) of the Adda-3 borehole compared to the gamma-ray and carbon- and oxygen-isotopic data (Perdiou et al., 2016). (B) Clay mineralogical data (this study) from Stevns-2 borehole compared to gamma-ray and carbon- and oxygen- isotopic data (Boussaha et al., 2016).

**Figure 3:** Stratigraphy and clay mineralogy of the Campanian in the Mons basin, Belgium and Campanian–Maastrichtian at El Kef, Tunisia. (A) Clay mineralogical data from the Cbr-7 borehole and carbonate content (this study) compared to carbon- and oxygen-isotope data and manganese contents (ppm) (Richard et al., 2005). (B) Clay mineralogical data from the El Kef – El Djebil section (this study), compared to carbon- and oxygen- isotopic data (Jarvis et al., 2002), carbonate content and Si/Al ratio (Mabrouk El Asmi, 2014). The foraminiferal biostratigraphic data of El Kef – El Djebil section are established from the lithostratigraphic comparison with Kalaat Senan section (see Jarvis et al., 2002), while the calcareous nannofossil biostratigraphic data have been performed on the El Kef – El Djebil samples. *G. aegyptica* = *Globotruncana aegyptica*; *G. elevata* = *Globotruncanita elevata*; *G. f.* = *Globotruncana falsostuarti*; *G. h.* = *Globotruncanella havanensis*; *G. g.* = *Gansserina gansseri*; *G. ventricosa* = *Globotruncana ventricosa*; *R. calcarata* = *Radotruncana calcarata*

*A. c. var NT = Arkhangelskiella cymbiformis; A. c. var W = Arkhangelskiella cymbiformis var W; B. p. c. = Broinsonia parca constricta;* C. spine = curved spine nannolith*; E. e. = Eiffellithus eximius; M. cf. M. p. = Micula cf M. premolisilva; R. a. = Reinhardites anthrophorus; T. o. = Tranolithus orionatus; U. g. = Uniplanarius gothicus; U. s. = Uniplanarius sissinghii; U. t. = Uniplanarius trifidius.*

**Figure 4:** Stratigraphy and clay mineralogy of the Campanian in the Umbria-Marche basin, Italy. Clay mineralogical data (this study) compared to carbon- and oxygen- data (this study) from (A) Gubbio – la Bottaccione and (B) Furlo – Upper Road sections. Magnetostratigraphy of Gubbio – la Bottaccione from Lowrie and Alvarez (1977); Furlo – Upper Road from Alvarez and Lowrie (1984). Gubbio biostratigraphic data from Coccioni and Premoli Silva (2015).

*A. m. = Archaeglobigerina minimus*; *D. a. = Dicarinella asymetrica*; *E. e. = Eiffellithus eximius*; *G. a. = Globotruncana aegyptica*; *G. e. = Globotruncanita elevata*; *G. h. = Globotruncanella havanensis*; *G. gansseri = Gansserina gansseri*; *G. st. = Globotruncanita stuarti*; *G. v. = Globotruncana ventricosa*; *R. l. = Reinhardites levis*; *R. m. = Rucinolithus magnus*; *U. g. = Uniplanarius gothicus*; *U. t. = Uniplanarius trifidus.*

**Figure 5:** Occurrence of selected calcareous nannofossils taxa in El Kef – El Djebil section.

**Figure 6:** Cross-plot of carbon- and oxygen-isotope bulk-rock data of the Gubbio – la Bottaccione and Furlo – Upper Road sections.

**Figure 7:** Cross-plot of carbon- and oxygen-isotope bulk-rock data of the Gubbio – la Bottaccione (▲) and Furlo – Upper Road (△) sections compared with isotopic data from several sites in the Tethyan Realm (A) to the north of 35 °N and (B) to the south of 35 °N.

**Figure 8:** Correlation of the $\delta^{13}C$ profiles across the Campanian between Adda-3 (Perdiou et al., 2015), Stevns-2 (Boussaha et al., 2016), Cbr-7 (Richard et al., 2005), Poigny borehole (Chenot et al., 2016), Tercis-les-Bains section (Voigt et al., 2012), Gubbio – la Bottaccione

section (this study), Furlo – Upper Road section (this study) and El Kef – El Djebil section (Jarvis et al., 2002).

**Figure 9:** Comparison of clay mineralogical assemblages of Campanian sediments of the sites studied (smectitic minerals are not represented). Correlation based on carbon-isotope events, along a palaeolatitudal transect from ~20°N to ~40°N. Adda-3 borehole, clay mineralogical data (this study) compared to the carbon- isotopic data (Perdiou et al., 2016); Stevns-2 borehole, clay mineralogical data (this study) compared to carbon- isotopic data (Boussaha et al., 2016); Cbr-7 borehole, clay mineralogical data (this study) compared to carbon- isotopic data (Richard et al., 2005); Poigny borehole, clay mineralogical data compared to carbon-isotopic data (Chenot et al., 2016); Tercis-les-Bains section, clay mineralogical data (Chenot et al., 2016) compared to carbon–isotopic data (Voigt et al., 2012); Gubbio – la Bottaccione, clay mineralogical data compared to carbon- isotopic data (this study); Furlo – upper Road section, clay mineralogical data compared to carbon- isotopic data (this study); El Kef – El Djebil section, clay mineralogical data (this study) compared to carbon – isotopic data (Jarvis et al., 2002).

**Figure 10:** Geodynamic framework of Campanian Tethyan bauxites and the clay minerals of the studied sections (modified from Bárdossy and Dercourt, 1990; Jolivet et al. 2016). Location of the sites studied: [A] Adda-3 borehole; [C] Cbr-7 borehole; [F] Furlo – Upper Road section; [G] Gubbio – la Bottaccione section; [K] El Kef – El Djebil section; [P] Poigny borehole; [S] Stevns-2 borehole; [T] Tercis-les-Bains section.

Name of the Campanian bauxites localities: (1) Alpilles (France), (2) Haut-Var (France), (3) Markusovce (Slovakia), (4) La Boissière (France), (5) Nurra (Sardinia), (6) Villeveyrac basin (France), (7) Bédarieux (France), (8) Tyrol Brandenberg and Salzburg (Austria), (9) Unterlaussa (Austria), (10) Sümeg (Hungary), (11) Halimba (Hungary), (12) Ihakut-Németbanya (Hungary), (13) Grméc Hill (Bosnia Herzegovina), (14) Jajce (Bosnia

Herzegovina), (15) Grebnik (Kossovo), (16) Kücük Koras, Sebimlkoy (Turkey), (17) Payas, Islaye (Turkey), (18) Sohodol, Cimpeni (Romania), (19) Euboea Island (Greece).

**Figure 11:** (A) Palaeolatitudinal topographic profile for the studied sites and their positions relative to the main orogenic belt during the studied time interval (see also Fig. 10) compared with (B) a reconstruction of the latitudinal climatic zonation with the locations of major bauxites. These provide a potential source of hydrolytic minerals into the adjacent sedimentary basin during the Santonian–Maastrichtian interval. (C) Palaeolatitudinal transect, from the base of the Late Campanian Event to the top of the end of late Campanian, of the mean clay minerals assemblages from sites studied in the Tethyan and Boreal realms. Location of the sites studied: [A] Adda-3 borehole; [C] Cbr-7 borehole; [F] Furlo – Upper Road section; [G] Gubbio – la Bottacione section; [K] El Kef – El Djebil section; [P] Poigny borehole; [S] Stevns-2 borehole; [T] Tercis-les-Bains section. There is no direct correspondence between A and B frames; the formation of bauxite and kaolinite occurred in humid tropical conditions. The synthetic "orogenic belt" (defined in A) represents both the Pyrenees and Alpine units that have their own geodynamic histories.

**Figure 12:** Scenario of the African block rotation modification, placed on a palaeogeographic map of the west Tethyan – south Boreal realm, from the end of the Santonian to the late Campanian. Histograms illustrate the proportion of clay mineral species (excluding the smectitic minerals background sedimentation) at the sites studied, during the (A) end Santonian, (B) early Campanian to early mid-Campanian, (C) later mid-Campanian, (D) Late Campanian Event, and (E) late Campanian.

# TABLES

**Table 1:** Peak positions used for the recognition of clay minerals (>2 µm) of the insoluble residue.

**Table 2**: Synthesis of carbon-isotope events recognised in the Campanian.

## SUPPLEMENTARY DATA

**Supplementary Data A:** Carbon and oxygen isotopes of the bulk rock, lithology and clay mineralogy of the Furlo – Upper Road section.

**Supplementary Data B:** Carbon and oxygen isotopes of the bulk rock, calcium carbonate content, lithology and clay mineralogy of the Gubbio – la Bottaccione.

**Supplementary Data C:** Carbon and oxygen isotopes of the bulk rock, calcium carbonate content, lithology and clay mineralogy of the Cbr-7 borehole.

**Supplementary Data D:** Carbon and oxygen isotopes of the bulk rock, lithology and clay mineralogy of the Stevns-2 borehole.

**Supplementary Data E:** Carbon and oxygen isotopes of the bulk rock, lithology and clay mineralogy of the Adda-3 borehole.

**Supplementary Data F:** Carbon and oxygen isotopes of the bulk rock, calcium carbonate content, lithology and clay mineralogy of the El Kef – El Djebil section.

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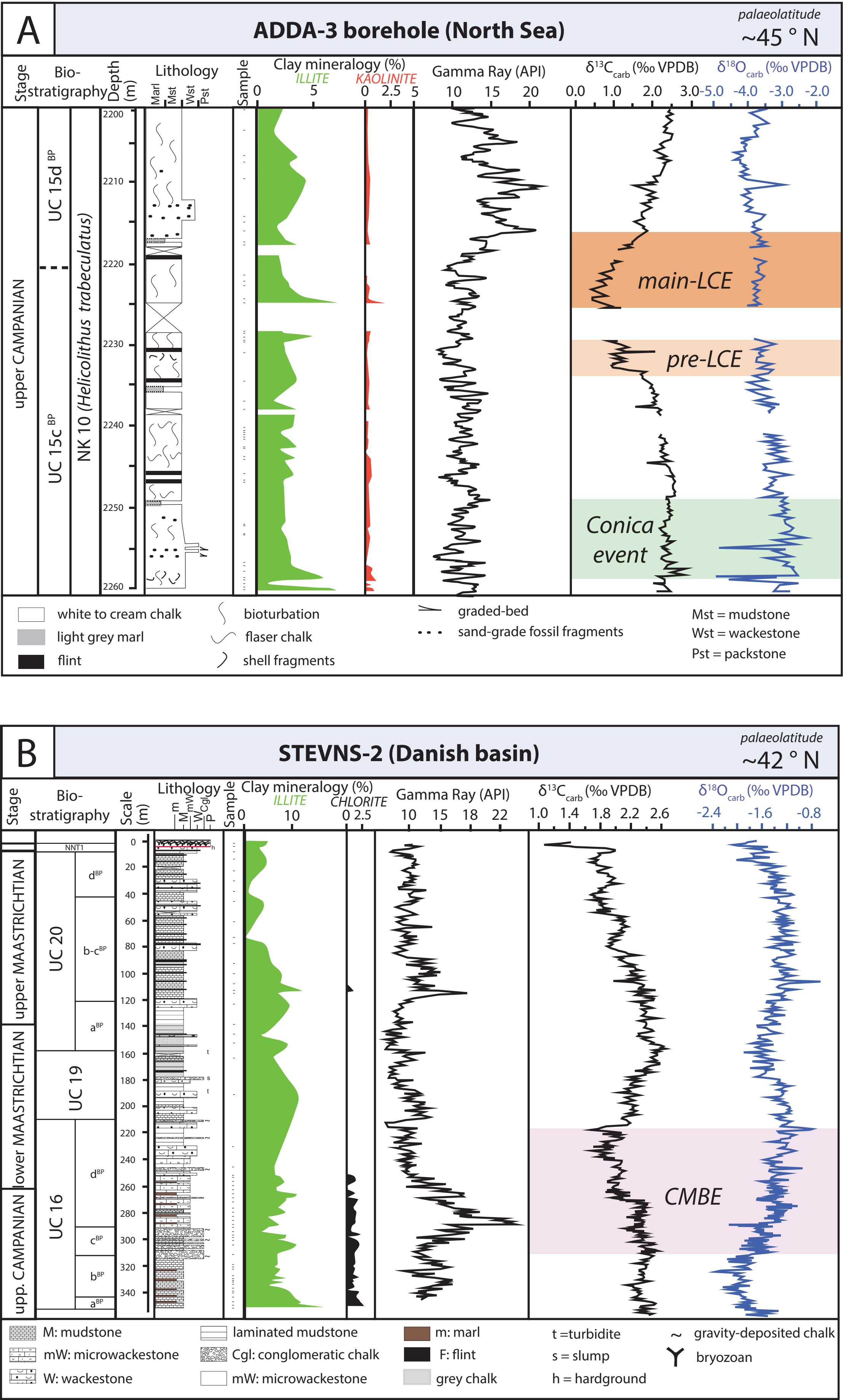
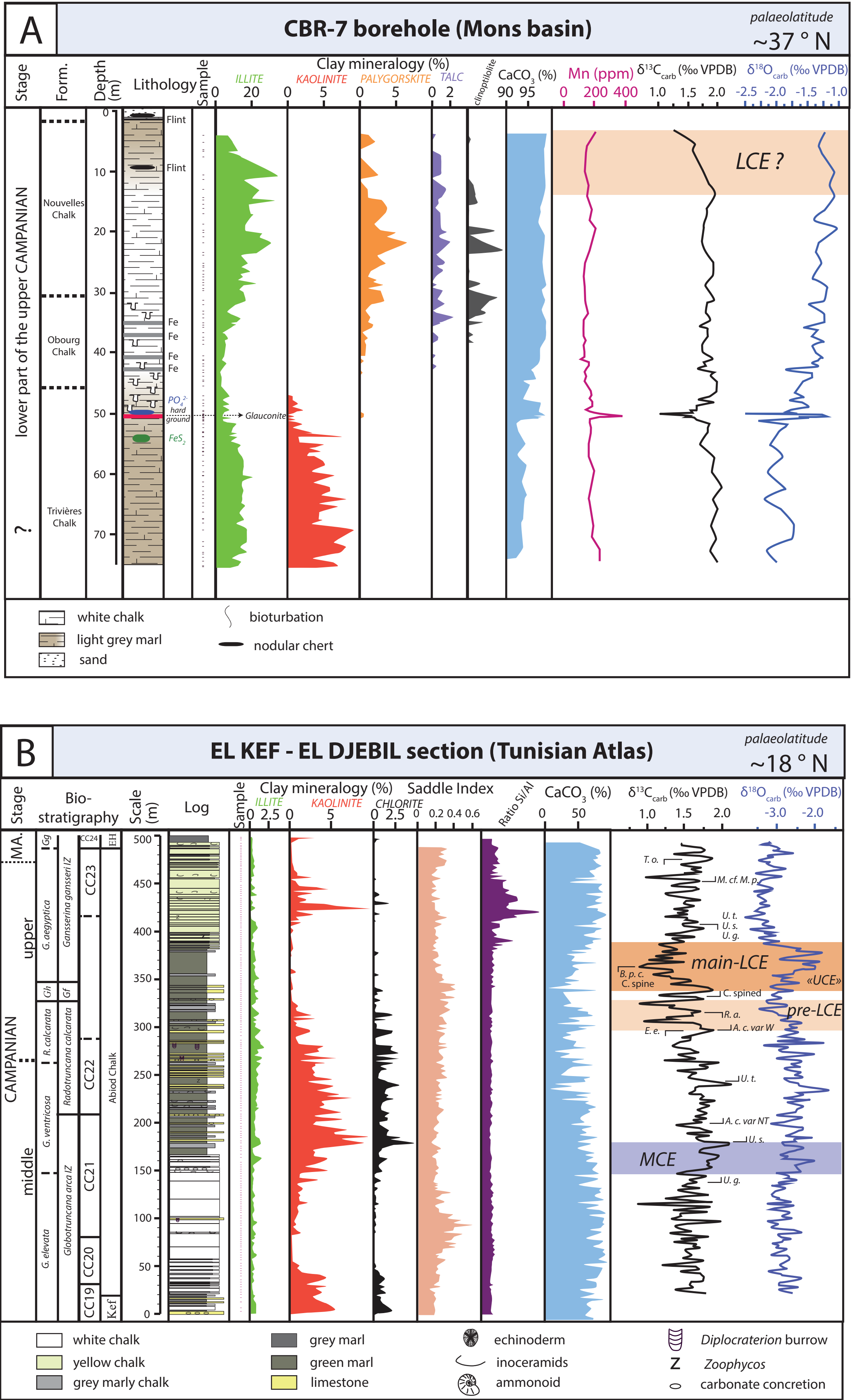
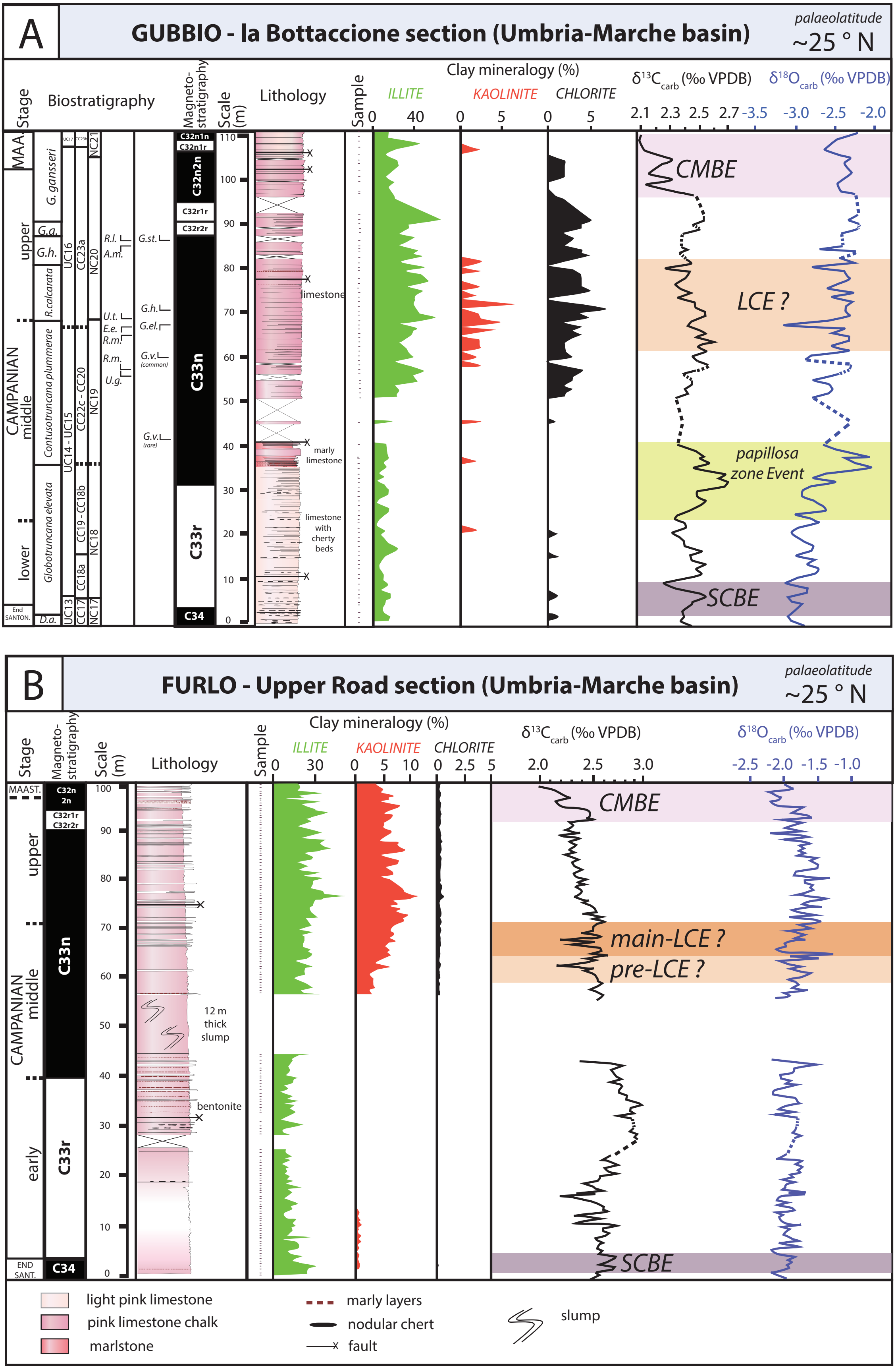
Grampian High
Irish Massif
Welsh Massif
A
S
Baltic Sea Basin
inverted Mid-Polish Anticline
40°N
Ukrainian Basin
Rhenish Massif
Bohemian Massif
C
P
Armorican Massif
Paris Basin
Silesia
Ukrainian Shield
Massif Central
Aquitaine Basin
Eastern Black Sea
30°N
T
Provence
Moesia
Corsica-Sardinia Block
Ebro Massif
High Karst
Western Black Basin
Iberian Massif
F
G
Dalmatia
ACP
Apulia
Ca
Kab
Ao
20°N
Jebilet High
Essaouira Basin
Atlasic Trough
K
Kas
Tarfaya Basin
Anti Atlas
Saharan Platform
Lybian Basin
Mardin
Aleppo Plateau
Tripolitaine
Alakhdar
Cyrenaic Platform
Tinrhet
Syrte Basin
Palmyrides
10°N
Rutbah
supposed coastlines
present coastlines of continents
normal fault
oceanic subduction
volcano
N
emerged land
fluviatile/lacustrine sediments
eolian sediments
coastal marine, shallow marine (terrigenous)
shallow marine carbonate deposits (platform, build-up and ramps)
deep(er) carbonate, (hemi)pelagic oozes
deep marine
0 250 500 km
new data sets
previously published data sets
ACP = Apennine Carbonate Platform
Ao = Alboran
Ca = Calabria
Kab = Kabylia
Kas = Kasserine Island



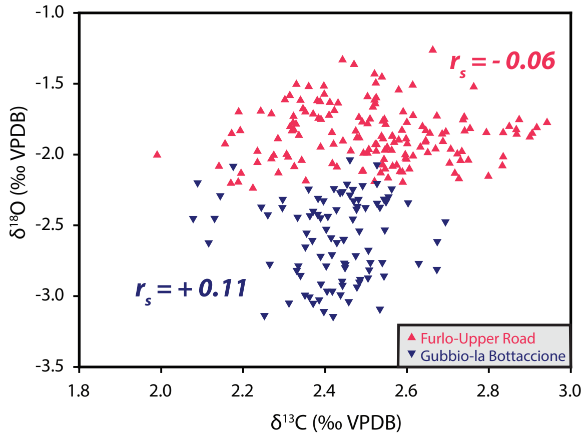
A
ADDA-3 borehole (North Sea)
palaeolatitude
~45 ° N
Stage
Bio-stratigraphy
Depth (m)
Lithology
Marl
Mst
Wst
Pst
Sample
Clay mineralogy (%)
ILLITE
KAOLINITE
Gamma Ray (API)
δ13Ccarb (‰ VPDB)
δ18Ocarb (‰ VPDB)
upper CAMPANIAN
UC 15d BP
UC 15c BP
NK 10 (Helicolithus trabeculatus)
main-LCE
pre-LCE
Conica event
white to cream chalk
light grey marl
flint
bioturbation
flaser chalk
shell fragments
graded-bed
sand-grade fossil fragments
Mst = mudstone
Wst = wackestone
Pst = packstone
B
STEVNS-2 (Danish basin)
palaeolatitude
~42 ° N
Stage
Bio-stratigraphy
Scale (m)
Lithology
m
M
mW
W
Cgl
P
Sample
Clay mineralogy (%)
ILLITE
CHLORITE
Gamma Ray (API)
δ13Ccarb (‰ VPDB)
δ18Ocarb (‰ VPDB)
NNT1
upper MAASTRICHTIAN
lower MAASTRICHTIAN
upp. CAMPANIAN
UC 20
UC 19
UC 16
dBP
b-cBP
aBP
cBP
bBP
CMBE
M: mudstone
mW: microwackestone
W: wackestone
laminated mudstone
Cgl: conglomeratic chalk
mW: microwackestone
m: marl
F: flint
grey chalk
t =turbidite
s = slump
h = hardground
gravity-deposited chalk
bryozoan


A
CBR-7 borehole (Mons basin)
palaeolatitude
~37 ° N
Stage
Form.
Depth (m)
Lithology
Sample
Clay mineralogy (%)
ILLITE
KAOLINITE
PALYGORSKITE
TALC
clinoptilolite
CaCO3 (%)
Mn (ppm)
δ13Ccarb (‰ VPDB)
δ18Ocarb (‰ VPDB)
lower part of the upper CAMPANIAN
?
Nouvelles Chalk
Obourg Chalk
Trivières Chalk
Flint
Fe
PO4 2-
hard ground
Glauconite
FeS2
LCE ?
white chalk
light grey marl
sand
bioturbation
nodular chert
B
EL KEF - EL DJEBIL section (Tunisian Atlas)
palaeolatitude
~18 ° N
Stage
Bio-stratigraphy
Scale (m)
Log
Sample
Clay mineralogy (%)
ILLITE
KAOLINITE
CHLORITE
Saddle Index
Ratio Si/Al
CaCO3 (%)
δ13Ccarb (‰ VPDB)
δ18Ocarb (‰ VPDB)
MA.
upper
middle
CAMPANIAN
Gg
G. aegyptica
Gh
R. calcarata
G. ventricosa
G. elevata
Gansserina gansseri IZ
Gf
Radotruncana calcarata
Globotruncana arca IZ
CC24
CC23
CC22
CC21
CC20
CC19
EH
Abiod Chalk
Kef
T. o.
M. cf. M. p.
U. t.
U. s.
U. g.
main-LCE
B. p. c.
C. spine
«UCE»
C. spined
R. a.
pre-LCE
E. e.
A. c. var W
A. c. var NT
MCE
white chalk
yellow chalk
grey marly chalk
grey marl
green marl
limestone
echinoderm
inoceramids
ammonoid
Diplocraterion burrow
Zoophycos
carbonate concretion

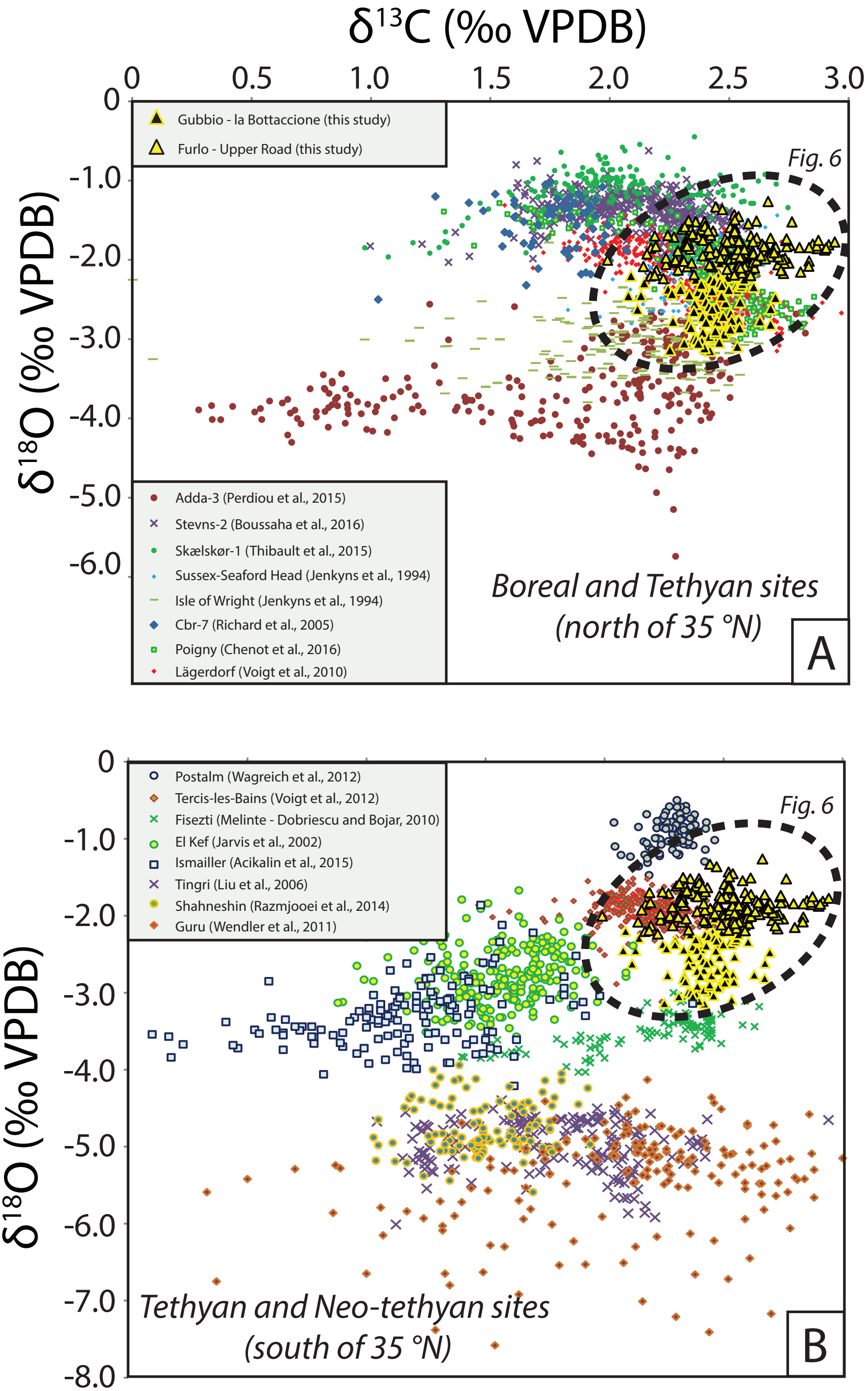

A
GUBBIO - la Bottaccione section (Umbria-Marche basin)
palaeolatitude ~25 ° N
Stage
Biostratigraphy
Magneto-stratigraphy
Scale (m)
Lithology
Sample
Clay mineralogy (%)
ILLITE
KAOLINITE
CHLORITE
δ13Ccarb (‰ VPDB)
δ18Ocarb (‰ VPDB)
MAA.
upper
CAMPANIAN middle
lower
End SANTON.
G. gansseri
G.a.
G.h.
R.calcarata
Contusotruncana plummerae
Globotruncana elevata
D.a.
UC16
UC14 - UC15
UC13
CC23a
CC22c - CC20
CC19 - CC18b
CC18a
CC17
NC21
NC20
NC19
NC18
NC17
R.l.
A.m.
U.t.
E.e.
R.m.
U.g.
G.st.
G.h.
G.el.
G.v. (common)
G.v. (rare)
C32n1n
C32n1r
C32n2n
C32r1r
C32r2r
C33n
C33r
C34
limestone
marly limestone
limestone with cherty beds
CMBE
LCE ?
papillosa zone Event
SCBE
B
FURLO - Upper Road section (Umbria-Marche basin)
palaeolatitude ~25 ° N
MAAST.
upper
CAMPANIAN middle
early
END SANT.
C32n 2n
C32r1r
C32r2r
C33n
C33r
C34
12 m thick slump
bentonite
CMBE
main-LCE ?
pre-LCE ?
SCBE
light pink limestone
pink limestone chalk
marlstone
marly layers
nodular chert
fault
slump

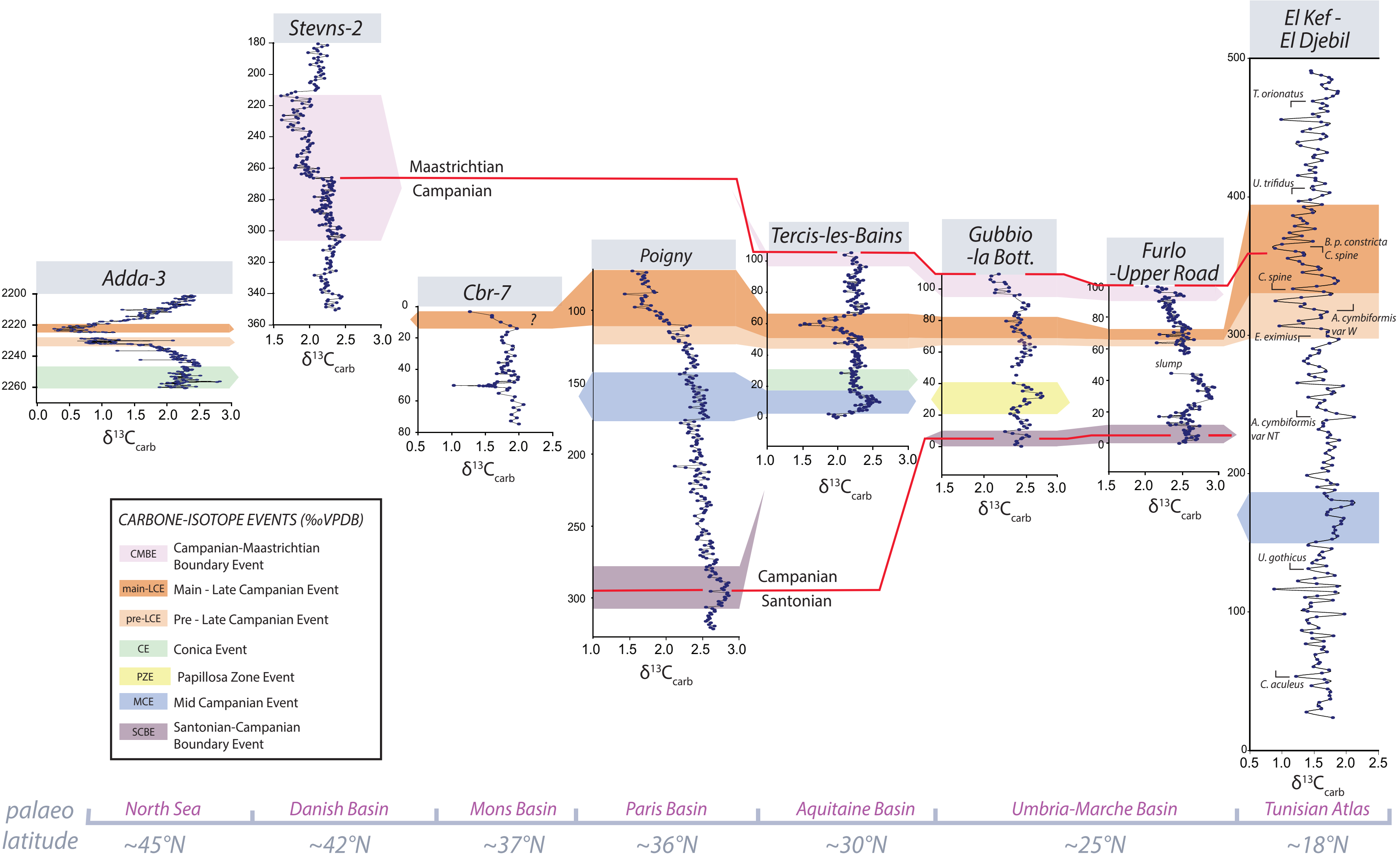
| Height (m) | Zeugrhabdotus bicrescenticus | Watznaueria barnesiae | Watznaueria biporta | Watznaueria manivita | Cribrosphaerella ehrenbergii | Prediscosphaera cretacea | Tranolithus minimus | Eiffellithus collis | Tranolithus gabalus | Chiastozygus cf. C. amphipons | Placozygus fibuliformis | Retecapsa crenulata | Micula staurophora | Gartnerago segmentatum | Rhagodiscus angustus | Retecapsa angustiforata | Rhagodiscus splendens | Microrhabdulus undosus | Staurolithites sp. | Zeugrhabdotus embergeri | **Calculites obscurus** | **Reinhardites levis** | Lithraphidites carniolensis | Micula sp. | Thoracosphaera sp. | Zeugrhabdotus sigmoides | Corollithion cf. C. signum | Rhagodiscus reniformis | **Eiffellithus eximius** | **Reinhardites anthophorus** | Biscutum constans | **Broinsonia parca constricta** | **Lucianorhabdus cayeuxii** | Eiffellithus casulus | **Tranolithus orionatus** | Cylindralithus sp. | **Ceratolithoides aculeus** | Ahmuellerella octoradiata | Manivitella pemmatoida | Eiffellithus turriseiffelii | Microrhabdulus decoratus | Helicolithus trabeculatus | Microrhabdulus belgicus | Zeugrhabdotus cf. Z. praesigmoides | **Uniplanarius gothicus** | Quadrum gartneri | Arkhangelskiella sp. | **Uniplanarius sissinghii** | Staurolithites mielnicensis | Arkhangelskiella cymbiformis var. NT | **Uniplanarius trifidus** | Lucianorhabdus maleformis | Kamptnerius magnificus | Lithastrinus grillii | Arkhangelskiella cymbiformis var. W | Curved spine | Watznaueria ovata | Prediscosphaera grandis | Eiffellithus parallelus | Micula cf. M. swastica | Micula cf. M. premolisilva | **Nannofossil biozonations (CC: zonation of Sissingh 1977 and Perch-Nielsen, 1985; UC: TP tethyan zonation of Burnett 1998** | | **sub-stage** | **STAGE** | **Main bioevents** |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 495,90 | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | | | | | | | | | | | + | + | + | + | + | + | | | | | + | | | + | | | | | | | | | + | + | + | **CC24** | **UC18** | **early** | **Maastri-chtian?** | |
| 474,60 | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | | + | | | | + | + | | + | | | | | | | | | | + | + | + | | + | + | | + | | | | | + | | | | | | | | | | | | | + | | **CC23b** | **UC17** | | | HO *T. orionatus* |
| 452,00 | + | + | + | | + | + | + | + | + | + | + | + | + | | | + | | + | | | | | | + | + | | | | | | | | | + | + | | + | | + | + | | + | | | | | | | | + | | | | | + | | + | | + | + | | | | | | |
| 434,2 | + | + | + | + | + | + | | + | + | | + | | + | | | + | | + | | | | | + | + | | | | | | | | | | | + | | + | | | | + | + | | | | | + | | | | | | | | | | + | | | | | | | | | |
| 403,00 | + | + | + | + | + | + | | + | + | + | + | + | + | | + | + | | + | | | | + | + | + | + | | | | | | | | | | + | + | + | | | | + | + | | | + | + | + | + | | | + | | | | | | | | | | | | **UC16a** | **late** | **Campanian** | HO *U. trifidius* |
| 367,80 | + | + | + | + | + | + | | + | + | | + | + | + | + | + | + | | + | | + | + | + | + | + | + | | | | | | | + | + | | + | | + | | | + | + | | | | + | + | + | | + | | + | | | | | + | | + | | | | **CC23a** | | | | HO curved spine, HO *B. parca constricta* |
| 335,3 | + | + | + | + | + | + | + | | + | + | + | | + | + | | | | + | | + | + | + | | + | + | | | | | | + | + | + | | + | | + | | + | + | + | + | | + | + | + | + | + | + | + | + | | | | + | + | + | | | | | | | | | LO curved spine |
| 316,80 | + | + | + | + | + | + | + | + | + | + | + | + | + | | | + | | + | | | + | + | | | + | | | | | + | + | + | + | | + | | | + | | + | + | + | | + | | + | | + | | + | + | | | | | | | | | | | **CC22** | **UC15d-e** | | | HO *R. anthophorus* |
| 299,3 | + | + | + | | + | + | + | + | + | + | + | + | + | + | | + | | + | | + | + | + | | + | + | + | | | + | | + | + | + | | + | | + | | | | | + | | | | | | + | + | + | + | | | + | + | | | | | | | | | | | HO *E. eximius* |
| 273,80 | + | + | + | | + | + | | | | | + | + | + | + | + | + | + | + | | + | + | | + | | | | | | | + | + | | + | + | + | | | + | + | + | | | | | + | | | | | | + | + | + | | | | | | | | | | | | | |
| 243,4 | + | + | + | + | + | + | | + | | + | + | + | + | | | + | + | + | + | + | + | + | | + | + | | | + | + | + | + | + | + | + | + | + | + | | + | | | | | + | + | | + | | + | + | + | | | | | | | | | | | | | | | LO *U. trifidus* |
| 203,85 | + | + | + | + | + | + | | | + | + | + | + | + | + | | + | + | + | | + | + | + | | | | | | | | + | | + | + | | + | | + | | + | + | | | | | + | + | + | + | + | + | | | | | | | | | | | | **CC21** | **UC15c** | | | |
| 174,1 | + | + | + | + | + | + | + | + | + | + | + | | + | + | + | + | | + | + | + | + | + | + | + | + | + | + | | | | + | | + | + | + | + | + | | | + | + | + | | | | | + | + | | | | | | | | | | | | | | | | | | |
| 135,30 | + | + | + | + | + | + | + | + | + | | + | | | + | | + | | + | | | | + | | + | + | | + | | + | | | + | | | + | | + | | | | + | + | + | + | + | + | + | | | | | | | | | | | | | | | **CC20** | **UC15b** | **early** | | LO *U. sissinghii* |
| 98,55 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| 55,9 | + | + | + | + | + | + | + | + | + | + | + | | + | + | | + | | + | | | + | + | + | + | | + | | | + | | + | + | + | + | + | | + | + | + | + | + | + | | | | | | | | | | | | | | | | | | | | | | | | LO *C. aculeus* |
| 2,2 | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | + | | | | | | | | | | | | | | | | | | | | | | | | | | **CC18-CC19** | **UC14b-UC15a** | | | |

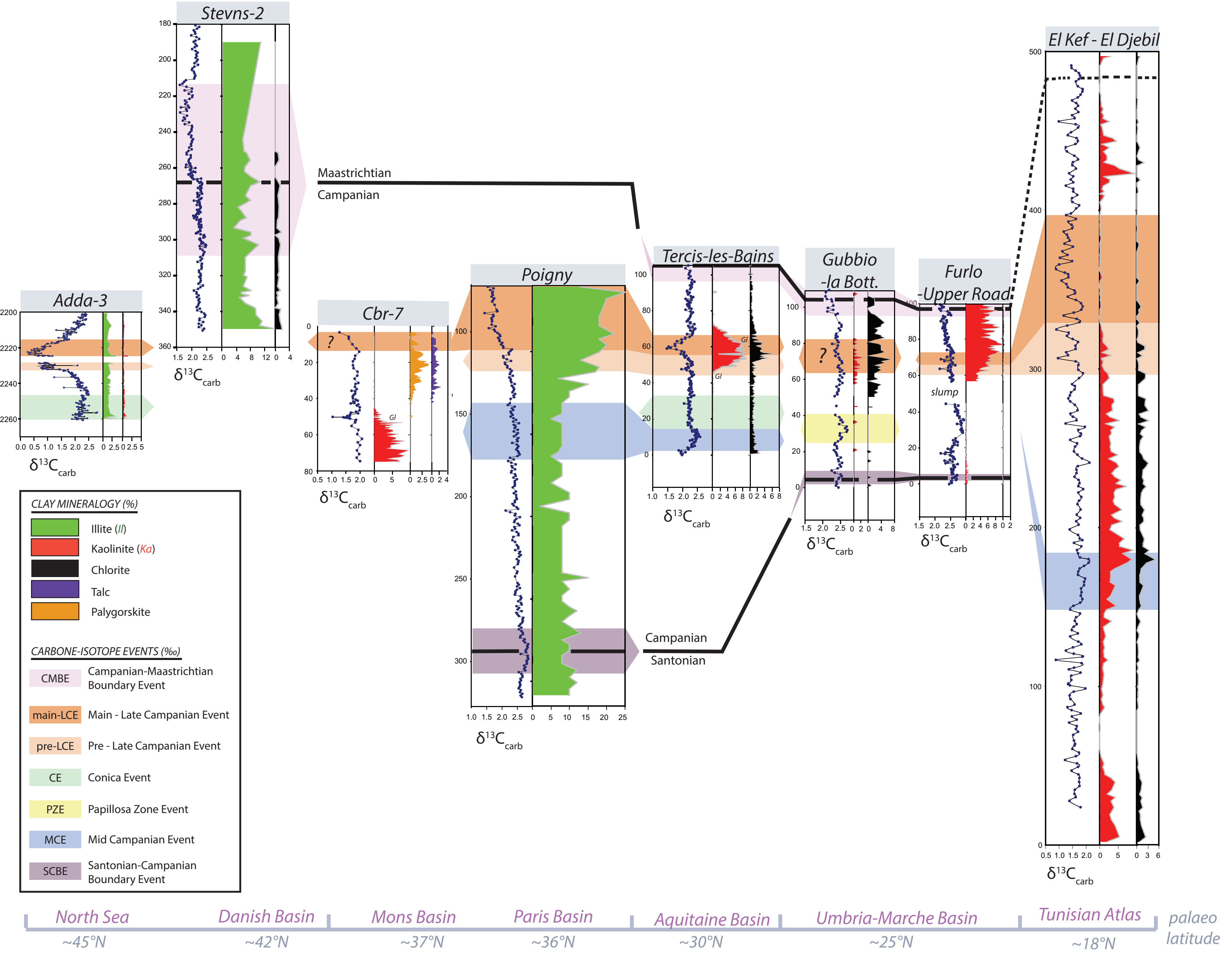
- + marks the presence of the species
- + First occurrence
- + Last occurrence
- highlights stratigraphic markers used for the biozonations



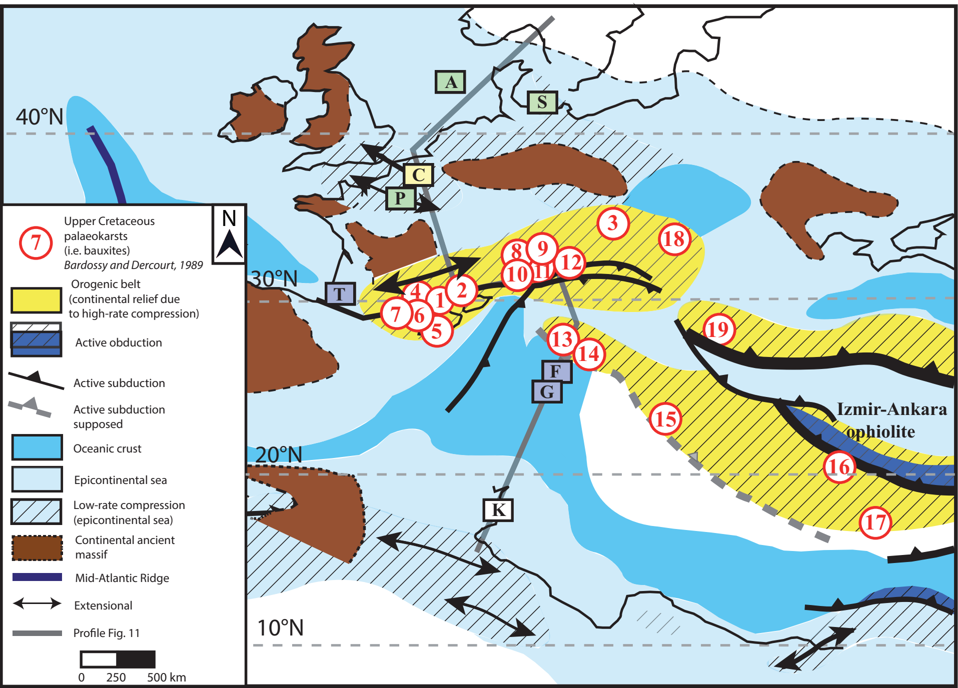
-1.0
-1.5
-2.0
-2.5
-3.0
-3.5
δ18O (‰ VPDB)
1.8
2.0
2.2
2.4
2.6
2.8
3.0
δ13C (‰ VPDB)
r s = - 0.06
r s = + 0.11
Furlo-Upper Road
Gubbio-la Bottaccione


δ13C (‰ VPDB)
0
0.5
1.0
1.5
2.0
2.5
3.0
δ18O (‰ VPDB)
0
-1.0
-2.0
-3.0
-4.0
-5.0
-6.0
Gubbio - la Bottaccione (this study)
Furlo - Upper Road (this study)
Fig. 6
Adda-3 (Perdiou et al., 2015)
Stevns-2 (Boussaha et al., 2016)
Skælskør-1 (Thibault et al., 2015)
Sussex-Seaford Head (Jenkyns et al., 1994)
Isle of Wright (Jenkyns et al., 1994)
Cbr-7 (Richard et al., 2005)
Poigny (Chenot et al., 2016)
Lägerdorf (Voigt et al., 2010)
Boreal and Tethyan sites
(north of 35 °N)
A
δ18O (‰ VPDB)
0
-1.0
-2.0
-3.0
-4.0
-5.0
-6.0
-7.0
-8.0
Postalm (Wagreich et al., 2012)
Tercis-les-Bains (Voigt et al., 2012)
Fisezti (Melinte - Dobriescu and Bojar, 2010)
El Kef (Jarvis et al., 2002)
Ismailler (Acikalin et al., 2015)
Tingri (Liu et al., 2006)
Shahneshin (Razmjooei et al., 2014)
Guru (Wendler et al., 2011)
Fig. 6
Tethyan and Neo-tethyan sites
(south of 35 °N)
B

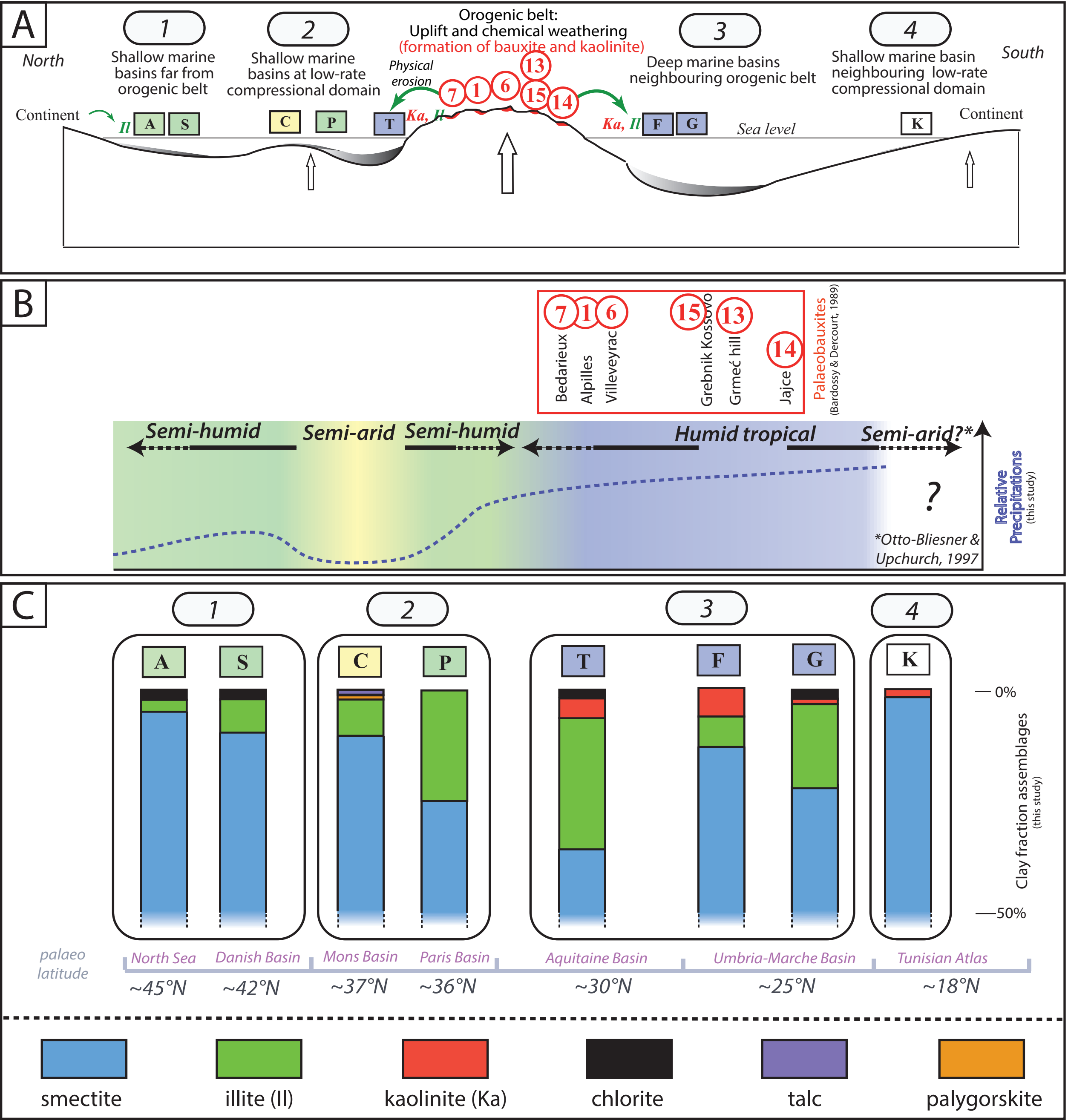

Stevns-2
Adda-3
Cbr-7
Poigny
Tercis-les-Bains
Gubbio
-la Bott.
Furlo
-Upper Road
El Kef -
El Djebil
Maastrichtian
Campanian
Campanian
Santonian
δ13Ccarb
slump
T. orionatus
U. trifidus
B. p. constricta
C. spine
C. spine
A. cymbiformis
var W
E. eximius
A. cymbiformis
var NT
U. gothicus
C. aculeus
CARBONE-ISOTOPE EVENTS (‰VPDB)
CMBE Campanian-Maastrichtian Boundary Event
main-LCE Main - Late Campanian Event
pre-LCE Pre - Late Campanian Event
CE Conica Event
PZE Papillosa Zone Event
MCE Mid Campanian Event
SCBE Santonian-Campanian Boundary Event
palaeo
latitude
North Sea ~45°N
Danish Basin ~42°N
Mons Basin ~37°N
Paris Basin ~36°N
Aquitaine Basin ~30°N
Umbria-Marche Basin ~25°N
Tunisian Atlas ~18°N

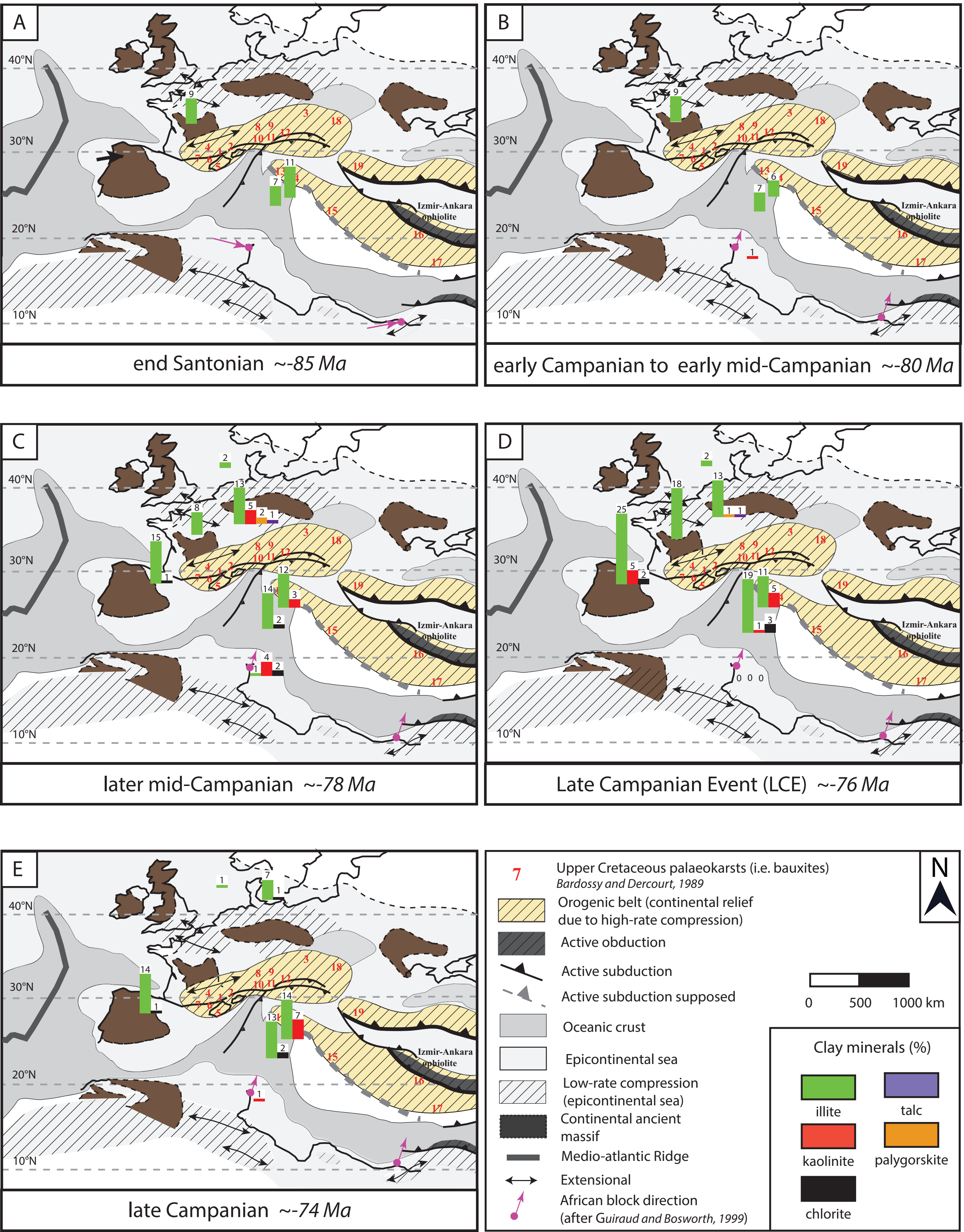


Stevns-2
Adda-3
Cbr-7
Poigny
Tercis-les-Bains
Gubbio
-la Bott.
Furlo
-Upper Road
El Kef - El Djebil
Maastrichtian
Campanian
Campanian
Santonian
slump
$\delta^{13}C_{carb}$
CLAY MINERALOGY (%)
Illite (Il)
Kaolinite (Ka)
Chlorite
Talc
Palygorskite
CARBONE-ISOTOPE EVENTS (‰)
CMBE
Campanian-Maastrichtian Boundary Event
main-LCE
Main - Late Campanian Event
pre-LCE
Pre - Late Campanian Event
CE
Conica Event
PZE
Papillosa Zone Event
MCE
Mid Campanian Event
SCBE
Santonian-Campanian Boundary Event
North Sea
Danish Basin
Mons Basin
Paris Basin
Aquitaine Basin
Umbria-Marche Basin
Tunisian Atlas
~45°N
~42°N
~37°N
~36°N
~30°N
~25°N
~18°N
palaeo latitude



40°N
30°N
20°N
10°N
A
S
C
P
T
F
G
K
Izmir-Ankara ophiolite
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
N
Upper Cretaceous palaeokarsts (i.e. bauxites)
Bardossy and Dercourt, 1989
Orogenic belt (continental relief due to high-rate compression)
Active obduction
Active subduction
Active subduction supposed
Oceanic crust
Epicontinental sea
Low-rate compression (epicontinental sea)
Continental ancient massif
Mid-Atlantic Ridge
Extensional
Profile Fig. 11
0
250
500 km


A
North
1
Shallow marine basins far from orogenic belt
2
Shallow marine basins at low-rate compressional domain
Orogenic belt: Uplift and chemical weathering
(formation of bauxite and kaolinite)
Physical erosion
3
Deep marine basins neighbouring orogenic belt
4
Shallow marine basin neighbouring low-rate compressional domain
South
Continent
Sea level
Ka, Il
B
Bedarieux
Alpilles
Villeveyrac
Grebnik Kossovo
Grmeć hill
Jajce
Palaeobauxites
(Bardossy & Dercourt, 1989)
Semi-humid
Semi-arid
Semi-humid
Humid tropical
Semi-arid?*
Relative Precipitations
(this study)
*Otto-Bliesner & Upchurch, 1997
C
0%
50%
Clay fraction assemblages
(this study)
palaeo latitude
North Sea
Danish Basin
Mons Basin
Paris Basin
Aquitaine Basin
Umbria-Marche Basin
Tunisian Atlas
~45°N
~42°N
~37°N
~36°N
~30°N
~25°N
~18°N
smectite
illite (Il)
kaolinite (Ka)
chlorite
talc
palygorskite


A
end Santonian ~-85 Ma
B
early Campanian to early mid-Campanian ~-80 Ma
C
later mid-Campanian ~-78 Ma
D
Late Campanian Event (LCE) ~-76 Ma
E
late Campanian ~-74 Ma
40°N
30°N
20°N
10°N
Izmir-Ankara ophiolite
7 Upper Cretaceous palaeokarsts (i.e. bauxites)
Bardossy and Dercourt, 1989
Orogenic belt (continental relief due to high-rate compression)
Active obduction
Active subduction
Active subduction supposed
Oceanic crust
Epicontinental sea
Low-rate compression (epicontinental sea)
Continental ancient massif
Medio-atlantic Ridge
Extensional
African block direction (after Guiraud and Bosworth, 1999)
N
0 500 1000 km
Clay minerals (%)
illite
talc
kaolinite
palygorskite
chlorite