Orbital calibration of the late Campanian carbon isotope event in the North Sea

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Abstract: A new record of carbon isotopes, nannofossil biostratigraphy, gamma-ray and Fe content variations is presented for the upper Campanian of the Adda-3 core, Danish Central Graben, North Sea. The studied interval provides a revision of previously assigned late Coniacian to early Santonian ages. New biostratigraphic data indicate a late Campanian age for the 60 m thick studied interval. The Late Campanian Event (LCE) is well recorded by a 1.5‰ negative excursion in the bulk δ13C, along with two stepwise pre-excursion negative shifts (defining the pre-LCE). The amplitude of the LCE appears higher in the North Sea than in other areas as seen from the correlation to Germany, the UK and France. This correlation allows identification of a new 0.4‰ negative excursion (defined as the conica event). Fe and gamma-ray variations are used to calibrate the record with cyclostratigraphy. Fourteen 405 kyr cycles identified in the upper Campanian of Adda-3 can be correlated to North Germany. The compilation of previous results from North Germany and correlation to Adda-3 shows that the Boreal upper Campanian spans a total of 17 cycles each of 405 kyr; that is, 6.885 myr. The duration of the LCE is estimated to be c. 1 myr at Adda-3 and in North Germany.

Supplementary materials: Calibration of the HH-XRF data is available at https://doi.org/10.6084/m9.figshare.c.2134362.

Received 3 June 2015; revised 15 October 2015; accepted 16 October 2015

Many studies have recently focused on the stratigraphy of the Maastrichtian stage and the Campanian–Maastrichtian boundary (CMB) (Husson et al. 2011; Batenburg et al. 2012, 2014; Gardin et al. 2012; Thibault et al. 2012a,b; Voigt et al. 2012; Batenburg 2013). These studies have considerably improved the geological timescale of the Maastrichtian with an astronomical calibration of the biostratigraphy and of high-resolution carbon isotope curves (Batenburg et al. 2012, 2014; Thibault et al. 2012a,b; Voigt et al. 2012). The same approach needs to be extended into earlier parts of the Late Cretaceous. Some calibrations of carbon isotopes and biostratigraphy to cyclostratigraphy have been proposed for large parts of the Late Cretaceous prior to the Maastrichtian (Buonoconto et al. 2002; Locklair & Sageman 2008; Voigt et al. 2010; Wagreich et al. 2012; Sprovieri et al. 2013; Sageman et al. 2014; Laurin et al. 2015). However, the Campanian stage lacks a proper calibration of the biostratigraphy and carbon isotope variations to cyclostratigraphy. Some of the best high-resolution carbon isotope records for this stage have been produced in the Boreal realm (Jenkyns et al. 1994; Voigt et al. 2010). High-resolution carbon isotope stratigraphy has been established for the Cenomanian–Campanian of the English Chalk, Coniacian–Maastrichtian of North Germany and late Campanian–Maastrichtian of the Danish Basin but the Late Cretaceous of the North Sea has been completely overlooked (Jarvis et al. 2001, 2002, 2006; Thibault et al. 2012a; Voigt et al. 2012). Here, we fill these gaps by presenting a new high-resolution carbon isotope record for the upper Campanian of the North Sea calibrated to cyclostratigraphy and calcareous nannofossil biostratigraphy. This new record is compared with results of previous studies, in particular with those for Lägerdorf–Kronsmoor (North Germany), for which a compilation is built to establish a standard geological timescale for the Boreal late Campanian.

The Adda-3 borehole, North Sea

The Adda-3 well is a hydrocarbon exploration well located in the Danish Central Graben in the westernmost part of the Danish offshore sector, about 250 km from the mainland (Fig. 1). The Chalk Group is represented by the Tor, Hod and Hidra Formations, which were cored between 2072.6 and 2301.2 m (6800–7550 feet). The Chalk Group was deposited in an extensive epeiric sea, which covered large parts of the European continent (e.g. Håkansson et al. 1974; Hancock 1975). This Chalk Sea resulted from rapid flooding of low-relief land during the Late Cretaceous and earliest Paleocene climatic optimum. In the Danish Central Graben, up to 1500 m of chalk were deposited during this c. 40 myr timespan (Surlyk et al. 2003; Vejbæk et al. 2007).

The Danish Central Graben is the southern branch of the North Sea Rift system. It is a complex of grabens that came into existence during Middle to Late Jurassic rifting events. The graben complex consists of generally NNW–SSE-trending half-grabens bounded by the Coffee Soil fault to the east, and by the Mid North Sea High to the west. Inherited Late Jurassic basin morphology persisted during the Early Cretaceous, but Late Cretaceous tectonic inversion (Vejbæk & Andersen 1987) changed the locations of depocentres dramatically and affected seafloor morphology. In uplifted areas, local erosion and long periods of non-deposition occurred, whereas in areas of subsidence, thick intervals of chalk accumulated. The Adda area was at the eastern margin of a Late Jurassic and Early to late Late Cretaceous depocentre before inversion, but at the western margin of a depocentre after the inversion (Vejbæk & Andersen 1987; Vidalie et al. 2014).

Lithology

The interval of interest, on the core and for samples acquired from the core, is 2200.8–2260.8 m (7220–7417 feet 4 inches) (Fig. 2).
The lithology consists of white chalk with sparse 20 cm thick flint bands and 10–20 cm thick marl intervals. No primary sedimentary structures are observed apart from two graded beds (each a few centimetres thick) near 2254 m. Most of the sedimentary succession is fully homogenized through pervasive bioturbation. Diagenetic features, similar to the flaser and lenticular chalk of Garrison & Kennedy (1977), are common. No core was recovered from the 2224.74–2228.44 m interval. Therefore, the succession was divided into a lower and an upper interval for the purpose of a cyclostratigraphic analysis based on Fe variations. Total gamma-ray values are given here in American Petroleum Institute units (API). Gamma-ray value was measured during the drilling and thus presents no gap through the succession. No analyses were conducted below 2260.82 m, where the core condition is relatively poor.

Methods

Calcareous nannofossil biostratigraphy

Thirty-five samples were selected for standard smear slides with an average resolution of one sample every 1.7 m in the cored sections. We used the standard smear-slide preparation as described by Bown & Young (1998) with the following modifications. Because the preservation of the nannofossils is generally poor in the samples, we tried to obtain the best possible preservation by gently scraping the chalk instead of crushing it in a mortar. Also, the ultrasonication step described by Bown & Young (1998) was omitted. The obtained solution was thoroughly stirred and smeared onto a cover-slip. Observations were made at a magnification of 1600× for a total of 200 fields of view. Semi-quantitative counts were performed in the following fashion: abundant (A) if, on average, more than 10 specimens could be observed in a field of view; common (C) if 1–10 specimens could be observed in each field; few (F) if one specimen or more could be observed in every 2–10 fields of view; rare (R) if, on average, only one specimen could be observed in 11–100 fields of view; very rare (VR) if, on average, only one specimen could be observed in 100–200 fields of view; single (S) if only one specimen was observed in the whole sample (Table 1). Preservation of the assemblage was estimated based on Roth (1978). The biozonations of Burnett et al. (1998) and Fritsen (1999) were applied (Fig. 3) and the taxonomic concepts of Perch-Nielsen (1985), Varol (1989) and Young & Bown (1997) were considered here.

Stable isotope geochemistry

A total of 273 samples were collected with an average sampling spacing of 20 cm. Analyses were carried out at the Department of Geosciences and Natural Resource Management, University of Copenhagen, using a Micromass Isoprime spectrometer (Ullmann 2013). The extraction of CO₂ was executed by reaction with anhydrous orthophosphoric acid at 70°C. The oxygen and carbon isotope values are expressed in per mille relative to the V-PDB standard reference. Reproducibility (2SD) is better than 0.16‰ for oxygen and 0.08‰ for carbon.

Fe concentration determination using HH-XRF data

Fe concentration was determined by hand-held X-ray fluorescence (HH-XRF) directly on the core using an Innov-X Olympus Delta Premium 6000 handheld HH-XRF analyzer with 4W 10/40 kV X-ray Rh tube and a large-area, high-performance silicon drift detector (SDD) as previously used and calibrated by us (Lenniger et al. 2014; Zhang et al. 2015). The HH-XRF system is equipped with the Geochem mode in Innov-X Delta PC software. The raw data expressed in counts per second were converted to concentrations in ppm, by calibration to a standardized metal alloy coin and fundamental parameters of the ‘Geochem’ mode in the embedded Innov-X Olympus Delta PC software where 131 certified standards have been used for calibration of the instrument. Furthermore, the HH-XRF results were tested and adjusted through internal calibration via measurement of certified standards PACS-2 and J-Do1 in the Sediment- and Aqueous-geochemistry Laboratory, Department of Geosciences and Natural Resource Management (IGN), University of Copenhagen (Ullmann 2013).
Fig. 2. Lithology, gamma-ray logging and resistivity of the complete Adda-3 borehole. Non-studied parts of the core are shaded in grey.
Care was taken not to measure stylolitic horizons, as these show marked iron enrichments with Fe/Al ratios well above the average shale of 0.55 (see Wedepohl 1991; Lenniger et al. 2014). The final data consist of 844 measurements made with an average resolution of 7 cm directly on the core, with 60 s counting time on each beam (10 and 40 kV) giving both light and heavy elements. Data acquired directly on cores have been compared with those acquired on powders for a set of 11 data points. The two datasets tend to correlate ($R^2 = 0.6959$). A number of HH-XRF data acquired on the core have been compared with the results of inductively coupled plasma mass spectrometry analysis of neighbouring sample powders. On average, a good correlation has been found between the two datasets, suggesting that overall Fe variations are faithfully reproduced by HH-XRF analysis.

**Gamma-ray logging**

The Adda-3 gamma-ray (GR) well log (Fig. 2) is represented by 1001 data points with an average spacing of 15.24 cm (½ foot). This log depicts the radioactivity of the formation, expressed in API. The well was completed in 1985 and a number of GR wireline tools were used. Schlumberger, which conducted the survey, lists its typical logging speed as 275 m h$^{-1}$ with duration of each measurement of 2 s, implying that the tool has moved by half a foot during one measurement (c. 15 cm). However, the cone of influence of the measurement by the probe is c. 40 cm. Taking into account the drift of the probe during one measurement, it is considered here that gamma-ray values are smoothed over a running mean of 40–60 cm. The interval of interest chosen for the study of gamma-ray variations is from 2194.56 to 2264.05 m (7200–7428 feet) and consists of 457 GR data points.

**Cyclostratigraphy**

Iron XRF measurements have been favoured for cyclostratigraphy in a number of studies as iron shows a higher signal-to-noise ratio and a better hole-to-hole consistency than other elements, thus allowing a more accurate high-resolution composite depth scale (Röhl & Abrams 2000; Evans et al. 2004; Westerhold et al. 2007). The stability of Fe XRF measurements makes it ideal to study potential Milankovitch climate cyclicity in pelagic sediments and it was thus chosen here for the cyclostratigraphic analysis along with gamma-ray variations. The total organic carbon content is generally very low in the Upper Cretaceous chalk and gamma-ray variations thus reflect mainly variations in the clay content (via Th and K, as also seen when comparing with Rb and Al from the HH-XRF analysis). Both gamma-ray and Fe variations should thus be sensitive to continental weathering variation forced by insolation (Westerhold et al. 2007). Data were de-spiked prior to the analysis at 3SD for removal of a few outliers with either very low or very high values. Very low values associated with the thick stylolite at 2215.35–2215.63 m were deleted (Fig. 4). Fe and gamma-ray data were linearly interpolated at 5 cm and 15 cm, respectively. Long-term trends were removed from the original signal of both data series using a robust LOESS smoothing weighted average over 63% of the total time-series (Fig. 4). Spectral analyses were performed using

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**Table 1. Distribution chart of calcareous nannofossils in the Adda-3 core**

| Sample ID | depth (m) | calcareous nannofossils | other nannofossils | foraminifera | calcareous algae | radiolarian algae | chitinozoa | microfossils |
|-----------|-----------|-------------------------|-------------------|-------------|----------------|----------------|-----------|-------------|
| 31425 | 300–315 | 30% | 20% | 30% | 10% | 10% | 5% | 5% |
| 31426 | 315–330 | 20% | 30% | 20% | 20% | 10% | 10% | 10% |
| 31427 | 330–345 | 30% | 15% | 20% | 20% | 15% | 10% | 10% |
| 31428 | 345–360 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31429 | 360–375 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31430 | 375–390 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31431 | 390–405 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31432 | 405–420 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31433 | 420–435 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31434 | 435–450 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31435 | 450–465 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31436 | 465–480 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31437 | 480–495 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |
| 31438 | 495–510 | 20% | 30% | 20% | 10% | 20% | 15% | 10% |

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the multi-taper method (MTM; Thomson 1982) with red noise estimates from Schulz & Mudelsee (2002) and rewritten for evenly spaced data using Matlab™ by Husson et al. (2014). The red noise Monte Carlo simulations are based on an AR1 autocorrelation model and allow for the estimation of confidence levels based on a $\chi^2$ statistical test performed on the theoretical spectrum of the red noise. To extract the potential cyclicities identified by the cyclostratigraphic analysis, we used Taner filters for high-precision extraction of specific astronomical frequency targets (Taner 2000).

Orbital tuning based on the identification of Milankovitch frequencies was used to provide a relative timescale. After tuning the time-series, the same analyses were performed again in the time domain with new red noise simulations.

**Results**

**Calcarenous nannofossil biostratigraphy**

Preservation of the nannofossil assemblages is generally poor, with a few samples presenting very poor preservation, but is moderate in the uppermost metres of the studied interval (Table 1). Species richness and total abundance (nannofossils per field of view; N/FOV) are generally low, and are slightly higher in some of the uppermost samples where preservation is better (Table 1).

Nevertheless, preservation does not prevent the recognition of a number of important biohorizons such as the first occurrences (FO) of *Reinhardtites levis* and *Prediscosphaera stoveri*, at 2249.07 m and 2220.14 m respectively, and a single occurrence (S) of *Orastrum campanensis* at 2203.73 m in a moderately preserved assemblage. The studied interval thus corresponds to the upper part of zone NK10 in the North Sea scheme and subzones UC15c-dBP in the Boreal scheme of Burnett et al. (1998) (Mortimer 1987; Varol 1991; Burnett & Whitham 1999). The FO of *Prediscosphaera stoveri* marks the base of UC15cBP subzone at 2220.14 m.

**Stable isotopes**

A cross-plot of bulk carbonate carbon and oxygen stable isotopes shows no significant trend that could attest to limited impact of diagenericity as diagenetically overprinted carbonate sediments often display a positive correlation between the two ratios (Jenkyns et al. 1995; Mitchell et al. 1997). This is mostly true for sections that comprise stratigraphic intervals with lithological changes between marly and less marly chalk. Here, most of the interval is dominated by relatively homogeneous mudstone chalk and the lack of correlation between carbon and oxygen stable isotopes cannot be considered as a reliable assessment of a lesser degree of diagenetic alteration. Based on the rather poor preservation of calcarenous nannofossils, some degree of alteration of the isotopic signals is expected. A number of data points show significantly negative oxygen isotope values (down to $-5.74‰$) and constant $\delta^{13}C$ values (Fig. 5). This could indicate a...
diagenetic overprint of $\delta^{18}O$ values through burial. However, the most negative values in $\delta^{13}C$ do not exhibit significantly lower values in $\delta^{18}O$ (Fig. 5). As shown in many studies, primary carbon-isotope trends are generally well preserved in the chalk, making them a powerful stratigraphic tool (Jenkyns et al. 1994; Jarvis et al. 2006; Voigt et al. 2010; Thibault et al. 2012). Although diagenetic alteration appears as a non-negligible feature of oxygen isotope values, it is likely that carbon-isotope trends can be used as a reflection of global changes in the carbon cycle.

Oxygen isotopes

The $\delta^{18}O$ values range between a minimum of $-5.74\%$o and a maximum of $-2.16\%$o, with a mean of $-3.65\%$o. Overall, values decrease upwards from $c. -3\%$ at the base of the studied interval to a minimum of $-4.5\%$ at 2207 m. Above this level, values increase upwards and reach $-3.5\%$ at the top (Fig. 6). In the lowermost part, there are four negative spikes, which most probably correspond to highly diagenetically altered horizons (Fig. 6). No anomalous values in $\delta^{13}C$ are recorded in association with these horizons.

Carbon isotopes

The $\delta^{13}C$ values range from 0.28 to 2.82‰ with a mean value of 1.81‰. A slight trend of increasing values is observed from 2260 to 2257 m (Fig. 6). This trend is interrupted by a very sudden 0.6‰ positive anomaly at 2257–2255.5 m followed by a sharp decrease and return to pre-excursion values. Values then fluctuate around a mean of 2.25‰ up to 2247 m. A sudden, sharp increase is observed at 2247 m up to values of 2.5‰. These two slight positive excursions centred at 2256 and 2247 m form the lower and upper shoulders of a characteristic slight, longer term negative excursion episode (Fig. 6). From 2247 m, values decrease progressively from 2.25‰ to 1.75‰ at 2235 m, after which a very sharp negative excursion of nearly 0.8‰ is recorded at 2232.3 m, followed by two additional cycles of stepwise negative excursions at 2229 m (1‰) and 2223.5 m (0.8‰). A minimum of 0.28‰ in $\delta^{13}C$ is recorded at 2222.5 m. The interval 2222.5–2215.0 m is characterized by a rapid recovery toward values of up to 1.8‰, and the rest of the studied interval shows a gentle progressive increase to 2.4‰ at the top (Fig. 6).
Fe concentration

The original Fe HH-XRF data (Fig. 4) present a minimum value of 75 ppm at 2206.52 m and a maximum value of 6284 ppm at 2248.92 m. The average value of the record is 2142 ppm. Iron can be present in a variety of forms but the most common form is in ‘clays’ (i.e. illite or smectite), oxy-hydroxides and pyrite. In general, the Fe/Al ratio is comparable with that of average shale, indicating no authigenic iron enrichment nor anoxic conditions (not shown). A few horizons have single point values elevated above a ratio value of unity, representing stylolite enrichments. The data illustrate a general declining trend in values upwards, implying either increased detrital input or a relative decrease of carbonate production for the lower part of the section (Fig. 4). Clear oscillations can be observed in the Fe HH-XRF record, which constitute the basis of the cyclostratigraphic analysis.

Cyclostratigraphy in the depth domain

The presence of long-term regular cycles can be seen by simple visual inspection of gamma-ray data, particularly in the upper part of the studied section (Fig. 2). The periodogram from the MultiTaper spectral analysis (MTM) of the gamma-ray signal highlights only one significant periodicity in a bandwidth between 4 and 6 m, and one peak with high power at 5.4 m (Fig. 7). Two peaks are present in the bandwidth centred around 1.4 m but they do not appear strikingly significant (Fig. 7). The MTM periodogram of iron concentration also shows significant peaks in the 4–6 m bandwidth of the upper and lower parts of the core (Fig. 7). In the MTM periodogram of iron concentration of the upper part of the core, two significant peaks are observed at 0.21 and 0.18 m. Less significant peaks are present at 1.2, 0.64, 0.35 and 0.29 m. In the lower part of the core, two significant peaks are observed at 0.22 m and 0.18 m. Less significant peaks are observed at 1.5, 1, 0.62 and 0.37 m.

Frequency ratios and cycle hierarchy of the peaks in the periodogram for iron suggest that the 4–6 m bandwidth corresponds to the 405 kyr eccentricity, peaks around 1–1.4 m correspond to the 100 kyr eccentricity band, peaks at 0.64–0.35 m correspond to the obliquity and peaks at 0.22–0.18 m correspond to the precession (Fig. 7). Filtering of the 405 kyr component that is well expressed both in the gamma-ray data and in Fe data was performed on a large bandwidth of 2.7–10 m in accordance with the bandwidth observed in MTM periodograms and to account for potential changes in sedimentation rates (Fig. 7). Filtering of the recognized 405 kyr eccentricity frequency bands in the lower and upper parts of the Fe time-series highlights about 14 cycles from 2200 to 2260 m, but their clear identification is hampered by the three core gaps of the studied record (Fig. 4). Filtering of the recognized 405 kyr frequency band in the gamma-ray data highlights 14 cycles from 1997.5 to 2260 m (Fig. 4). However, the phase relationship between the filtered Fe and gamma-ray series is unclear. Maxima and minima of these two filters are not in phase and the amplitude of many cycles in the filtered Fe series is often low. For this reason and because the gamma-ray actually has a continuous signal throughout the studied interval, an age-model based on 405 kyr cycles was derived from the filtered gamma-ray record only.

Maxima of the gamma-ray filter were chosen to define the boundaries between the 405 kyr cycles, except for the maximum at the top of UCa8, which was more difficult to locate and was selected halfway between two minima (UCa1–UCa10, Fig. 4). The identification of UCa10 may be subject to discussion because this cycle is shorter than any other and the gamma-ray filter does not exhibit a well-marked minimum at 2217 m. However, the 405 kyr filter of Fe data shows a better defined minimum at 2217.4 m and condensation in this interval is suspected owing to the presence of
two intervals with numerous stylolites at 2215.6 and 2219.4 m
(Fig. 4). Because of its poorly significant expression, the 100 kyr
eccentricity was not filtered here.

**Cyclostratigraphy in the time domain**

Extraction and identification of the 405 kyr eccentricity cycles from
the gamma-ray data allowed for building an age-model, which was
used to tune the time-series into the time domain. This exercise is
particularly useful for a re-analysis of cyclostratigraphy so that the
expression of potential Milankovitch cyclicities in a power
spectrum can be directly compared with that of the La2004
astronomical solution (Laskar et al. 2004; Fig. 7). The MTM
periodogram of the gamma-ray tuned time-series reveals an
enhanced expression of the 405 kyr. Three main peaks cross the
95% confidence level at 110, 93 and 78 kyr and probably
correspond to e2, e1 and e3 of the La2004 astronomical solution
(Fig. 7). Of these peaks, those at 93 and 78 kyr reach the 99%
significance level although the power of the three eccentricity
components is rather low as compared with that of the 405 kyr
(Fig. 7). When using identical tuning on the Fe series the MTM
power spectra also reveal significant Milankovitch frequencies.
The time-series of the upper part reveals peaks at 360 (E), 136 (e2),
52 (o2), 38 (o1), 25.5 (o3?), 23.5 (p1) and 18 kyr (p2) whereas the
time-series of the lower part reveals peaks at 385 (E), 128 (e2), 64
(o2?), 38 (o1), 32 (o3), 21.5 (p1) and 17 kyr (p2). Comparison of
these power spectra with the MTM power spectrum of La2004
argues for a correct identification of Milankovitch frequencies in the
Adda-3 record (Fig. 7).

**Discussion**

**Gamma-ray signal constraint to 405 kyr frequency peaks**

The stratigraphic ‘reach’ of the gamma-ray tool typically means that
one measurement integrates radiation from c. 40 cm (vertically) of
rock although the power of the radiation decreases toward the edges

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![Graph](https://via.placeholder.com/150)

**Fig. 5.** $x$–$y$ plot for bulk carbonate $\delta^{13}C$ and $\delta^{18}O$ data measured in the
Adda-3 core. The correlation coefficient is 0.277.

![Graph](https://via.placeholder.com/150)

**Fig. 6.** Carbon and oxygen stable isotopes records of the studied interval in Adda-3
with two important calcareous nanofossil biohorizons allowing biozonation of the
core.
Despite the sampling step of c. 15 cm, effectively, the gamma-ray tool performs a 40 – 60 cm near running mean smoothing of the rock radiation as explained in the Methods section. In the most condensed sections of the Adda-3 interval, the average rate of accumulation appears to be a little more than 1 cm ka$^{-1}$. Consequently, the smoothing of the GR suppresses the expression of the 20 and 40 kyr variations, and potentially weakens the expression of the 100 kyr eccentricity band. The nature of GR data sampling explains the absence of any significant peaks in the precession and obliquity bands and the rather low power of the short-eccentricity band as compared with the powerful expression of the 405 kyr component (Fig. 7). It is likely that here the full expression of the eccentricity band is slightly compromised by the smoothing of the gamma-ray signal. Therefore, the enhancement of the expression of the 78 kyr component as compared with the 93 and 110 kyr components is most probably a bias reflecting the nature of GR data acquisition.

**Stratigraphy of the Boreal upper Campanian**

The Cretaceous Stage Boundaries symposium held in Copenhagen in October 1983 was a major breakthrough in Cretaceous stratigraphy with the erection of the first comprehensive definition of Valanginian to Maastrichtian stage boundaries (Birkelund et al. 1983, 1984). Sections in NW Europe played a particularly important role in these definitions, supported by decades of work on macrofossil biostratigraphy. Very useful regional to supra-regional biostratigraphic markers were recognized, particularly among the groups of the echinoderms, inoceramids, belemnites and ammonites, with the first comprehensive attempts of correlation to benthic foraminifer and calcareous nanofossil biohorizons (Bailey et al. 1984; Birkelund et al. 1984; Kennedy 1984; Schulz et al. 1984). It was accepted that the extinction level of the free-living crinoid *Marsupites testudinarius* should be chosen as the best possible criterion for defining the base of the Campanian because of the nearly global distribution of this species (Schulz et al. 1984).

Before the ratification of the Campanian–Maastrichtian Global Stratotype Section and Point (GSSP) boundary at Tercis-les-Bains (SE France), the base of the Maastrichtian stage was assigned to the FO of belemnite *Belemnella lanceolata* with reference to the chalk section of Kronsmoor, North Germany (base of the *B. lanceolata* zone noted *lan.* in Fig. 8; Birkelund et al. 1984; Schulz et al. 1984; Schönfeld et al. 1996). However, the biogeographical distribution of this species is limited to the Boreal realm (Odin 1996). The Maastrichtian Working Group reassigned the base of the Maastrichtian to the FO of ammonoid *Pachydiscus neubergicus*, which has a much wider geographical distribution (Hancock 1991; Odin 1996), although it is characterized by only rare findings in the Boreal realm (Niebuhr & Esser 2003). The CMB was defined and ratified at the GSSP of Tercis-les-Bains close to the first occurrence of *P. neubergicus* (preferred guide event) and as the arithmetical mean of 12 distinct biohorizons (Odin & Lamoureille 2001). Niebuhr & Esser (2003) were the first investigators to establish that the Boreal definition of the CMB at Kronsmoor is not coincident with that of the GSSP. Rare findings of *P. neubergicus* in the Kronsmoor section allowed them to suggest that *B. lanceolata* first appeared c. 540 kyr earlier than *P. neubergicus* and that the base of the Maastrichtian as defined at Tercis-les-Bains should actually be placed at Kronsmoor at the top of the *Belemnella pseudobtusa* zone (noted *p.* in Fig. 8; see also Niebuhr 2006). More recently, carbon-
isotope stratigraphy allowed for an even more precise definition of the CMB, as the Late Campanian to early Maastrichtian interval is characterized by a global $0.4\,–\,1\,‰$ large and long-lasting negative excursion in the $\delta^{13}C$ record called CMBE (Voigt et al. 2010, 2012; Thibault et al. 2012a, b). The $\delta^{13}C$ negative shift that characterizes the onset of the CMBE has been further defined in three major steps at Stevns-1, Denmark, which has the highest resolution record for this excursion (Thibault et al. 2012a). CMBa corresponds to the first sharp negative shift, CMBb to a small positive rebound and CMBc to a second sharp negative shift. These three steps are more or less equivalent to steps CMBE1, CMBE2 and CMBE3 of Voigt et al. (2012) and can also be identified at Kronsmoor (Fig. 8). Thibault et al. (2012b) and Voigt et al. (2012) have shown that the level of the CMB as defined at Tercis-les-Bains corresponds to the base of CMBc (top of C32n2n of Voigt et al. 2012; Fig. 9), which correlates with the base of the $B$. obtusa zone at Kronsmoor (Fig. 8). The best calcareous nannofossil marker for this boundary appears to be the FO of Prediscosphaera mgayae (Thibault et al. 2012a, b, 2015; Sheldon et al. 2014).

Subdivisions of the Campanian have not been officially ratified and different concepts are used in different parts of the world. In the Boreal realm, the Campanian is traditionally subdivided in two substages, with the last occurrence (LO) of the Goniotholithus genus (LO $G$. quadrata gracilis) defining the base of the upper Campanian as well as the base of the conica–senior zone at Lägerdorf (noted co/se in Fig. 8 and named from belemnite Belemnella mucronata senior and echinoid Echinochorys conica, which have concurrent ranges in this zone; Schulz et al. 1984). In the UK, the base of the Upper Campanian is also defined by the LO of $G$. quadrata gracilis (Wood & Mortimore 1988). It is worth mentioning here that the FO of Belemnella mucronata considered as the former marker for the Boreal upper Campanian in the UK (Brydone 1912) and used in the...
Geological Time Scale 2012 (GTS2012, Ogg et al. 2012) is situated slightly below that level, at the base of the gracilis–senior zone (noted gr/se in Fig. 8). Ogg et al. (2012) mentioned that the latter stratigraphic biohorizon would project slightly below the middle–lower Campanian boundary in North American usage (Cobban et al. 2006). Niebuhr (2006) chose to subdivide the North German record in a lower, middle and upper Campanian by positioning the middle–lower Campanian boundary at the base of the gracilis–senior zone and the middle–upper Campanian boundary at the base of the polyplacum zone in the Kronsmoor section (noted polypl. in Fig. 8). The FO of the ammonoid Trachyscaphites spiniger, traditionally used for defining the lower–upper Campanian boundary in Poland and Spain, is situated in NW Germany one zone higher in the lower part of the basiplana–spiniger zone (bas/spin in Fig. 8) and correlates with the FO of calcareous nannofossil Reinhardtites levis (Schulz et al. 1984; Schönfeld et al. 1996). In this study, we chose to follow the upper Campanian definition of Schulz et al. (1984), with its lower boundary defined by the LO of G. quadrata gracilis equivalent to the base of the conica–senior zone at Lägerdorf (Fig. 8).

Correlation of biostratigraphy and carbon isotope stratigraphy to other upper Campanian records

The negative δ13C excursion situated immediately above the void at Adda-3 correlates well with the Late Campanian Event (LCE) as defined by Jenkyns et al. (1994) and Jarvis (2006) in the English chalk and also identified in North Germany (Voigt et al. 2010) and at Tercis-les-Bains, SE France (Voigt et al. 2012; Figs 8 and 9). This δ13C event is defined as a rather symmetrical negative excursion in the latter sections whereas it presents a strong asymmetry in Adda-3 with a high amplitude of the upper positive shift (Figs 8 and 9). However, it is likely that the basal part of the excursion is truncated in the Adda-3 record owing to the void (Figs 8 and 9). This isotopic correlation is supported by calcareous nannofossil biostratigraphy, which clearly indicates a late Campanian age for the studied interval. Conversely, the solid biostratigraphic assignment to late Campanian age of the studied interval of Adda-3 is further confirmed by the presence of the LCE. This age assignment is in strong contrast to the original biostratigraphic interpretation proposed for the entire core, which suggested a late Coniacian to early Santonian age (Fig. 2). Correlation of δ13C between Adda-3 and Lägerdorf–Kronsmoor is constrained by the FO of Reinhardtites levis in the lowermost part of the upper Campanian and the presence of Orastrum campanensis in the record from Adda-3, which last occurs in the upper Campanian of Lägerdorf–Kronsmoor (Fig. 8). Prediscosphaera stoveri first occurs slightly above the FO of R. levis in Lägerdorf–Kronsmoor whereas its FO is recorded within the LCE in Adda-3 (Fig. 8). Correlation between Adda-3 and Trunch (Norfolk, UK) shows a slightly less discrepant FO of P. stoveri between the two records (Fig. 9). The two sharp negative shifts in δ13C noted 1 and 2 in Figure 8 are interpreted here as two pre-LCE excursions. These two excursions can be correlated with a rather high confidence to the Lägerdorf–Kronsmoor record (Fig. 8). The correlation of the two excursions to Tercis-les-Bains and Trunch remain elusive although both sections express at least one clear pre-LCE excursion (Fig. 9). Taking into account the total stratigraphical range comprising the LCE and the pre-LCE excursions, the total amplitude of δ13C variations appears much greater in Adda-3 (c. 2‰) than in Trunch (Norfolk, UK) (c. 1‰), in Lägerdorf–Kronsmoor (North Germany) (c. 1‰) and in Tercis-les-Bains (SE France) (c. 1‰). The amplitude of δ13C variations thus seems to vary with location. This observation could reflect local processes affecting the Adda-3 carbon isotope record such as enhanced diageneric alteration enhancing the expression of pre-LCE excursions 1 and 2.
The top of the studied interval in Adda-3 is constrained by the correlation of δ13C to Lägerdorf–Kronsmoor. In Adda-3, the δ13C record above the LCE shows a gentle progressive increase whereas values remain fairly stable above the LCE in Lägerdorf–Kronsmoor. However, δ13C values also tend to increase slowly above the LCE at Norfolk (Fig. 9). The uppermost Campanian has been characterized globally by a rapid negative shift >0.5‰, which is part of the larger stepwise CMB negative excursion (Thibault et al. 2012a,b, Voigt et al. 2012). This first negative shift, defined as CMBa, can be observed in nannofossil zone UC16 in Lägerdorf–Kronsmoor but is not observed in Adda-3, suggesting that the top of the Adda-3 record lies somewhere below CMBa (Figs 8 and 9). Correlation of the top of the Adda-3 record is further constrained by the presence of Eiffelithus eximius throughout the studied interval in the core. The LO of E. eximius in Lägerdorf–Kronsmoor that marks the top of UC15 is in the middle of 405 kyr cycle UCa14. Correlation of carbon isotope curves between Lägerdorf–Kronsmoor and the Stevns-1 borehole in the Danish Basin confirms the reliability of this bio-event in the Boreal realm (Thibault et al. 2012a,b, 2015) and constrains the top of Adda-3 to the level of the LO E. eximius in North Germany, somewhere below 405 kyr cycle UCa15 (Fig. 8).

Additionally, the slightly negative excursion with sharp bracketing shoulders identified at the base of the Adda-3 δ13C record (between 2257 and 2247 m) may be correlated to similar excursions at Lägerdorf–Kronsmoor, Trunch and Tercis-les-Bains (Figs 8 and 9). This small excursion seems to be characteristic and useful for further correlations, we propose here to name this excursion the ‘conica event’ because it correlates to the base of the conica–senior macrofossil zone (as defined by the LO of G. quadrata gracilis) in North Germany (Fig. 8).

**Orbital calibration of the LCE and of the Boreal Late Campanian**

The cycles in the Adda-3 record can be numbered and appear to match well the Lägerdorf–Kronsmoor cycles identified by Voigt & Schönfeld (2010; Fig. 8). Minima in the 405 kyr filter of %CaCO3 data in Lägerdorf–Kronsmoor correlate to maxima of the 405 kyr filter extracted from our gamma-ray data (Fig. 8). It is possible to assign the numbering of upper Campanian 405 kyr cycles in the Adda-3 record, using the correlation of Adda-3 to Lägerdorf–Kronsmoor (UCa1–UCa14 in Figs 8 and 9). Calcareous nannofossil biostratigraphy and carbon isotope stratigraphy constrain the top of the Adda-3 record to somewhere below 405 kyr cycle UCa15 (Fig. 8). In addition, the correlation of the Adda-3 record is well anchored at the base by the newly defined conica event, which spans UCa1 and UCa2 in North Germany (Fig. 8). Therefore, options for correlation between the two δ13C records are bound at the base and top and the most negative δ13C values of the LCE can be used as a third anchor for this correlation (Fig. 8). With regard to these assumptions, counting of the 405 kyr cycles in Adda-3 and correlation to the 405 kyr cycles derived from the Lägerdorf–Kronsmoor record allows the recognition of cycles UCa1 to UCa14 in the gamma-ray filter whereas the top of the interval studied for biostratigraphy and δ13C reaches the top of UCa13 (Fig. 8).

Biostratigraphic and chemostratigraphic correlations of Adda-3 to North Germany show that the North Sea record comprises most of the upper Campanian. Additionally, the cyclostratigraphic age-model proposed by Voigt & Schönfeld (2010) on Lägerdorf–Kronsmoor is confirmed, allowing for an orbital calibration of the upper Campanian δ13C record and macrofossil zones (Fig. 8). The age-model proposed here is further confirmed by the correlation of the LCE, pre-LCE excursions and conica event at Adda-3 to Trunch (Fig. 9). Based on new cyclostratigraphic and radiometric constraints for the base of the Campanian in the Western Interior (Sageman et al. 2014), a calibration in time of the Trunch δ13C curve has been recently proposed by Laurin et al. (2015; Fig. 9). Taking an age of 72 Ma for the base of the first 405 kyr cycle of the Maastrichtian (Thibault et al. 2012a,b), δ13C curves of Adda-3 and Lägerdorf–Kronsmoor can be calibrated in age from the downward counting of 405 kyr cycles (Fig. 8). This calibration is in agreement with that of Laurin et al. (2015; Fig. 9). Consequently, the Boreal upper Campanian comprises 17 eccentricity cycles each of 405 kyr duration, accounting for a total duration of 6.885 myr. Voigt & Schönfeld (2010) proposed a duration of 6.2 myr for this interval using the former Boreal definition of the CMZ at the base of the lanceolata zone, thus truncating cycle UCa17 and half of UCa16 (Fig. 8). Adding these truncated cycles to their estimate provides the same duration as given above.

The total duration of the LCE is difficult to estimate properly in the Adda-3 record as it lacks the basal part of the event. The LCE in Adda-3 spans cycles UCa10, UCa9 and the upper part of UCa8, each 405 kyr in duration. Correlation to Lägerdorf–Kronsmoor shows a rather similar result, with the LCE spanning almost entirely UCa10, UCa9 and the upper half of UCa8 (Fig. 8). Consequently, the duration of the LCE can be estimated to be of the order of 1 myr.

At Adda-3, the overall large negative trend in δ13C is characterized by four main steps (Fig. 8):

1. a 0.8‰ slow progressive decrease commencing just above the conica event spans three and a half 405 kyr eccentricity cycles (i.e. c. 1400 kyr from the middle of UCa3 to the top of UCa6);
2. the two sharp negative excursions defined here as pre-LCE excursions 1 (0.8‰) and 2 (c. 1‰) span cycle UCa7 and the lowermost UCa8 (i.e. c. 600 kyr);
3. the LCE, recorded here by a c. 0.8‰ negative shift followed by a 1.5‰ positive recovery, spans the uppermost part of UCa8 to the top of UCa10 (i.e. c. 1 myr);
4. a slow 0.6‰ progressive increase spans at least three 405 kyr cycles (i.e. c. 1200 kyr from the base of UCa11 to the top of UCa13).

Interestingly, pre-LCE excursions 1 and 2 match fairly well the timing of UCa7 and UCa8 in Adda-3 and Lägerdorf–Kronsmoor, suggesting that the environmental perturbation responsible for the expression of the LCE first amplified the forcing of the carbon cycle by the 405 kyr eccentricity before its full expression during the LCE. Furthermore, at Lägerdorf and in Adda-3, sharp negative shifts in carbon isotopes can be systematically observed close to the base of UCa5 and UCa6, further confirming the Milankovitch forcing of carbon isotope variations in this overall interval (Fig. 8).

Pacing of the carbon cycle by Milankovitch cycles has been observed in many other Phanerozoic studies (Sprovieri et al. 2013; Laurin et al. 2015, and references therein). However, the relationship between carbon isotope variations and 405 kyr eccentricity cycles is less clear at times of significantly larger and long-lasting excursions such as during the LCE and the conica event. The larger amplitude and the long-term expression of the LCE and pre-LCE excursions in the Adda-3 record suggest that perturbations of the carbon cycle may have been amplified in the North Sea area as compared with the rest of the Boreal realm. However, we cannot exclude that this observation is due to diagenetic alteration and, if not, the exact cause of this amplified signal in the North Sea remains to be explained.

**Conclusions**

New investigations of the calcareous nannofossil biostratigraphy and carbon isotope stratigraphy of the upper Adda-3 core (Danish Central Graben, North Sea) allow for a significant revision of the
studied interval to a late Campanian age assignment in contrast to the previously considered late Cenomanian to early Santonian age. A large 1.5% negative excursion in δ^{13}C represents the late Campanian event (LCE) as constrained by the biostratigraphy and is preceded by two sharp pre-excursion negative shifts that suggest an amplification of the 405 kyr eccentricity pacing of the carbon cycle just prior to the LCE. The LCE and the pre-LCE excursions are correlated with the records of Lägerdorf–Kronsmoor (North Germany), Trunch (Norfolk, UK), and Tercis-les-Bains (SE France).

A small 0.4% negative δ^{13}C excursion at the base of the Adda-3 carbon isotope record can also be correlated to North Germany, Tercis-les-Bains and Trunch, and is defined here as the ‘conica event’ because of its correlation with the conica–senior macrnofossil zone in North Germany.

The cyclostratigraphy based on Fe and gamma-ray variations has highlighted frequency peaks identified as the precession, the obliquity, the short-eccentricity and the 405 kyr eccentricity.

Extraction of the 405 kyr eccentricity band from the gamma-ray signal highlights 14 cycles used to temporally tune the series. These cycles, together with the carbon isotope record, correlate well with previous results obtained at Lägerdorf–Kronsmoor (Voigt & Schönfeld 2010; Voigt et al. 2010). Ages derived from the cyclostratigraphy of Adda-3 and correlation to Lägerdorf–Kronsmoor point to the Boreal upper Campanian spanning a total of 17 cycles of each 405 kyr (i.e. 6.858 myr).

The duration of the LCE is of the order of 1 myr as constrained by cyclostratigraphy in Adda-3 and North Germany, whereas pre-LCE events correspond to well-marked 405 kyr cycles.

The amplitude of the LCE in Adda-3 (1.5%) is higher than at Lägerdorf–Kronsmoor, Trunch and Tercis-les-Bains (c. 1%). Further investigation is needed to explain the cause of this carbon cycle perturbation and its amplified signal in the North Sea.

Acknowledgements and Funding

This study was inspired by previous work of S. Voigt, whom we warmly acknowledge for original datasets of Lägerdorf–Kronsmoor (Voigt & Schönfeld 2010; Voigt et al. 2010). Ages derived from the cyclostratigraphy of Adda-3 and correlation to Lägerdorf–Kronsmoor point to the Boreal upper Campanian spanning a total of 17 cycles of each 405 kyr (i.e. 6.858 myr).

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Scientific editing by Quentin Crowley

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