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# Conjunctive use of spectral gamma-ray logs and clay mineralogy in defining late Jurassic–early Cretaceous palaeoclimate change (Dorset, U.K.)

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## Abstract

Detrital clay mineralogy is controlled by weathered source rock, climate, transport and deposition that in turn influence the spectral gamma-ray (SGR) response of resultant sediments. Whilst a palaeoclimate signal in clay mineralogy has been established in some ancient successions, the SGR response remains contentious, largely because the data sets have yet to be collected at the same or appropriate vertical scales to allow comparison. In addition, the influence of organic matter on SGR is not always considered. Here, we present clay mineralogical, total organic carbon (TOC) and SGR analyses from the late Jurassic and early Cretaceous of the Wessex Basin, a period of previously documented palaeoclimate change. The aim of this paper is to estimate the sensitivity of SGR as palaeoclimatic tool, SGR and clay mineral data having been collected at the same sample points, making this one of the most rigorous comparison of clay mineral and SGR to date. Overall, the correlation between high thorium/potassium or thorium/uranium and kaolinite associated with a well-established palaeoclimate change shows that elevated thorium may be used as a proxy for humid palaeoweathering, as suggested by few previous studies. © 2005 Elsevier B.V. All rights reserved.

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## 1. Introduction

Previous attempts to relate the response of SGR data to palaeoclimates has relied on the suggestion by

Rosholt (1992), Osmond and Ivanovich (1992) and Parkinson (1996) that the mobility of potassium (K) and uranium (U) and the relative concentration of thorium (Th) during weathering should result in clays with elevated Th/K and Th/U ratios. Thus Ruffell and Worden (2000) made a simple test of SGR response to a known period of palaeoclimate

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change, i.e. the Aptian interval, with some generally positive results. This work has yet to be tested against direct measurements of another associated proxy indicator of changing palaeoweathering conditions.

The Jurassic–Cretaceous boundary beds of England, Germany and France have long been known to contain clear evidence of a change from semi-arid to semi-humid climates (Allen, 1998), where plant fossils, spore-pollen, clay minerals, sedimentology and other data support the theory of arid to humid conditions. The late Jurassic and early Cretaceous semi-arid phase (early Tithonian–middle Berriasian) is followed by a wetter phase after the middle-late Berriasian (Allen, 1998). This pattern is widespread and has been recorded on the northern margin of Tethys, as well as on the southern margin and Atlantic realm, based on clay minerals (Hallam, 1984; Deconinck, 1993; Allen, 1998; Daoudi and Deconinck, 1994; Schnyder et al., 2005), as well as on spore-pollen data (Abbink et al., 2001). The climate change has also been recorded on the Russian Platform (Ruffell et al., 2002). The palaeogeographical limit of the climate change are, however, still not known. For example, using clay mineral data, the climate change seems not recorded in East Greenland (Lindgreen and Surlyk, 2000). This could constitute a northern limit of the Jurassic–Cretaceous boundary climate change. Nevertheless, Hallam et al. (1991), Weissert (1989) and Sellwood and Price (1993) all suggest that the widespread and long-lived changes in clastic vs. carbonate deposition documented in the Jurassic–Cretaceous of Europe have a palaeoclimatic origin. Carbonate and evaporite associations are thought to reflect semi-arid climates with clastic and coal associations indicating raised humidities, each of which are reflected in variations in clay mineralogy.

### 1.1. Outline

The aim of our work is to test the SGR theory by directly comparing our bed-by-bed clay mineral record of environmental change through the Jurassic–Cretaceous of southern England against SGR data. Additional data on the organic carbon content of the rocks are also used in order to control the possible influence of organic matter on SGR measurements.

Our work aimed to provide a half meter to meter-scale sample suite of clay mineralogy as the main

proxy indicator of palaeoclimate change (Chamley, 1989; Velde, 1995), that could test whether the SGR data that we obtained showed a commensurate change to both previously published and newly analysed indicators of palaeoclimate. Palaeoclimate signals are so often masked in the rock record we ensured that the depositional and diagenetic environments were well-established before commencing our work.

### 1.2. Mineralogy and burial

The clay mineral content of a typical Mesozoic mudstone from the study area is controlled by the type of weathered parent material, the weathering regime, the depositional environment and later diagenetic alteration. Diagenesis constitutes a general serious problem in interpreting SGR data. Another problem is possible concentrations of U- or Th-rich heavy minerals in sandy layers. In the two studied locations, we thus have chosen to compare clay mineralogy to SGR logs, using mainly mudstones and argillaceous limestone samples, previous studies (Westhead and Mather, 1996; Allen, 1998) having suggested that the shallow-buried and homogenous mudrock successions show limited clay mineral diagenesis. In the more heterolithic successions, diagenesis is largely confined to the porous rocks, which were avoided here. The successions have been buried to no more than 1000 m. Under the geothermal conditions experienced in NW European sedimentary basins, this would lead to a burial temperature of no more than 45–50 °C. Such temperatures are too low to cause major diagenetic reactions in mudrocks (Hardy and Tucker, 1988). Rock-Eval pyrolysis data (see below) also suggest immature or slightly mature character of organic matter ( $T_{\max}$  mainly below 435–440 °C) and therefore limited burial.

## 2. Geological setting

### 2.1. Wessex Basin

The studied area is located in the Dorset part of the Wessex Basin, one of the extensional sedimentary subbasins that covered much of NW Europe during the Mesozoic. At the Jurassic–Cretaceous boundary, active extension, faulting and crustal subsidence

occurred, starting in the late Jurassic, at the Oxfordian–Kimmeridgian boundary, and decreasing in the early Cretaceous, in Hauterivian–Barremian times (Chadwick, 1986). This led to a complex pattern of subsidence and uplift associated with tilted blocks, filled out by syn-rift sediments in the so-called Purbeck and Wealden facies (Chadwick, 1986; Underhill, 2002).

## 2.2. Lulworth and Durlston Formations (Purbeck Limestone Group: Jurassic–Cretaceous boundary)

The thinly bedded limestones, mudrocks, and evaporites, devoid of ammonites, that comprise the Purbeck Limestone Group in the Wessex Basin were deposited in shallow marine and non-marine environments at or near the sea-level, at the end of the Jurassic and during the earliest Cretaceous (West, 1975; Clements, 1993; Westhead and Mather, 1996; Batten, 2002). The Purbeck Limestone Group lie on a basal unconformity affecting the upper part of the Tithonian open-marine and ammonite bearing underlying unit (Allen and Wimbledon, 1991), and is overlain by early Cretaceous non-marine Wealden clastic facies. In the Dorset area, evaporitic environments interpreted as sabkha (West, 1975), lagoonal, lacus-

trine brackish and freshwater environments are recorded, with highly fluctuating salinities, between freshwater and hypersaline water, being indicated by molluscan, ostracod, charophyte and palynomorph distributions (reviews in Milner and Batten, 2002). Episodes of contemporaneous emersion occur throughout the sections and are indicated by dinosaurs' footprints, mudcracks, suggesting that there were no significant water depth changes. The shallow coastal environments were cut off most of the time from the open sea. However, several marine invasions are recorded, even in some freshwater/brackish environments, probably indicating the breaching or overflowing of coastal barriers.

We collated the pertinent lithostratigraphic data for the sections at Durlston Bay and Lulworth Cove (Figs. 1 and 2) (Clements, 1993; Westhead and Mather, 1996). The Durlston Bay Purbeck-type section is about 107 m thick and Lulworth Cove 32 m. The two sections are separated by about 20 km. The Lulworth Cove section is equivalent to the basal part of the Durlston Bay-type section, correlation between outcrops being classically based on marker-beds such as the Mammal Bed (M.B.) and the Cinder Bed (C.B.) (Fig. 2, see below). In the absence of ammonites, age-assignment of the Durlston Bay sec-

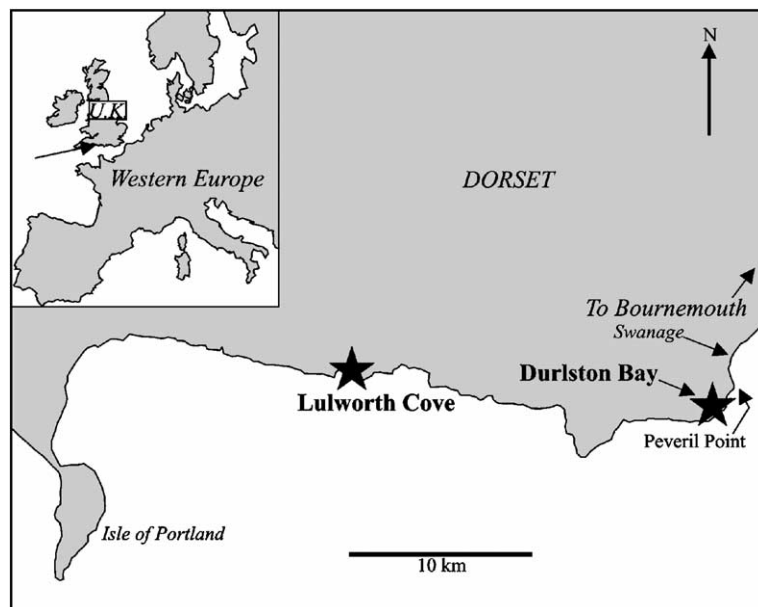
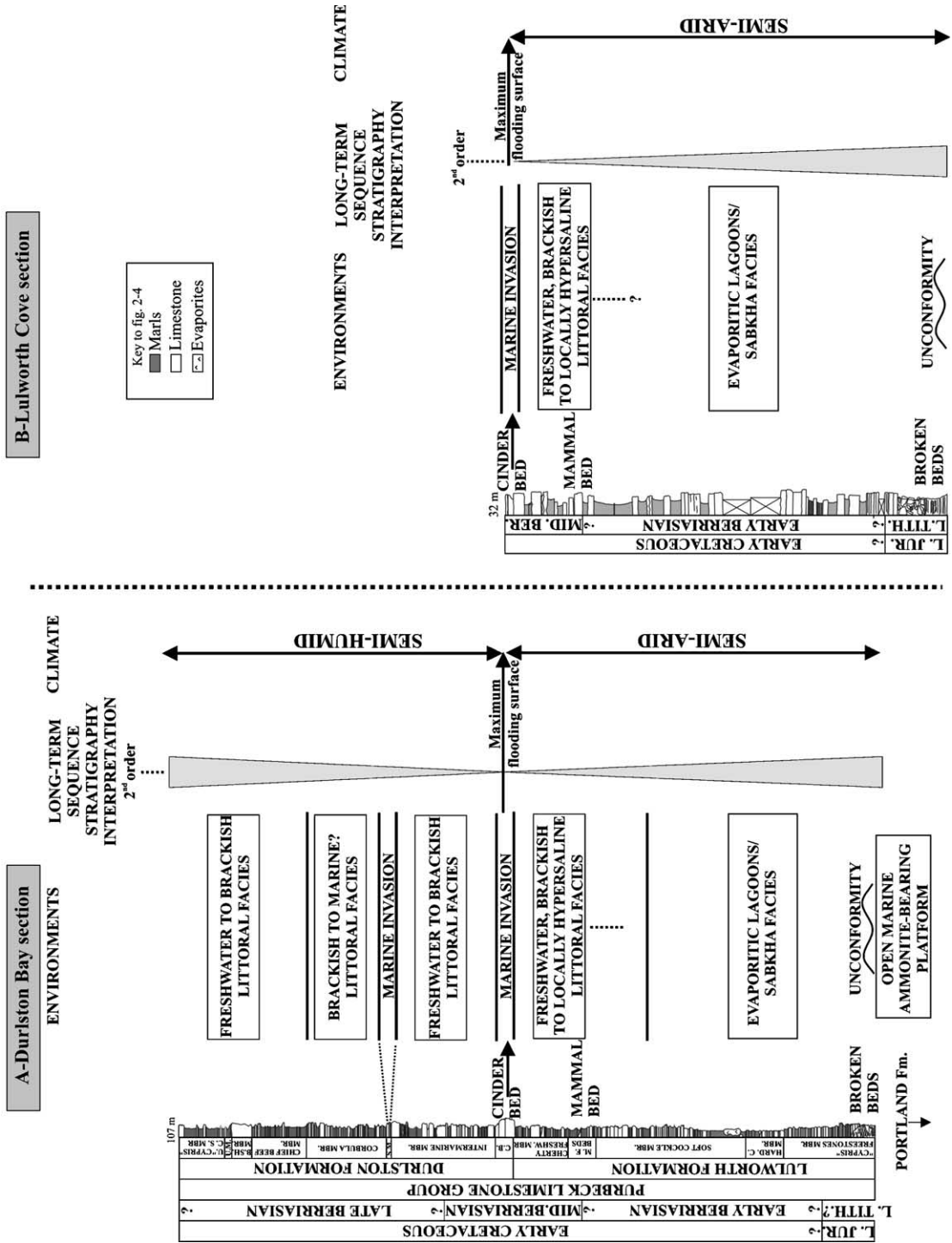


Fig. 1. Location map of the studied sections in the Dorset area, southern England.



tion is based on charophytes (Feist et al., 1995), palynology (Abbink et al., 2001), and magnetostratigraphy (Ogg et al., 1994). The Durlston Bay section probably represents most of the Berriasian stage. A comprehensive outcrop and drilling survey demonstrates the lateral continuity of the main beds throughout Dorset (Westhead and Mather, 1996), allowing use of selected sections for studies such as this. In this study, we have followed the lithostratigraphic scheme of Clements (1993), who subdivided the section into two formations (Lulworth and Durlston beds) and thirteen informal members (see text below), together with bed numbers (also used here). Fig. 2A–B summarizes the lithostratigraphy, together with the trends in changing salinities recorded by Radley (2002).

### 2.3. Sea-level changes

The facies succession, at a large-scale, can be interpreted in terms of fluctuating relative sea-level, following the European-scale sequence stratigraphic scheme of Jacquin et al. (1998). The basal unconformity of the Purbeck Limestone Group is linked to a major late-Jurassic sea-level fall (sequence boundary (SB) Ti7 or Ti8, Jurassic–Cretaceous unconformity, Jacquin et al., 1998) (Fig. 2A–B). The general regressive trend reaches its maximum above the studied interval, in the Wealden Group, where extensive freshwater to brackish environments are recorded (late Berriasian–Valanginian). Together with localized uplift, the general regression lead to an extensive erosion pattern, especially in Wealden facies (Chadwick, 1986).

Within this general trend, the occurrence (in the middle part of the Purbeck Group), of the widespread Cinder Bed, a massive brackish to near-marine shelly

limestone, with occasional urchin spines, indicates the main marine incursion recorded in these coastal facies. A more minor marine transgression is recorded above, in the Scallop Member (Fig. 2A). However, it has been suggested that Purbeck–Wealden brackish/marine episodes could not be truly transgressive, but the result of localized sedimentary events (Allen, 1998). In addition, some high frequency salinity fluctuation appear to be mostly the result of runoff/evaporation balance and not of sea-level changes (Radley, 2002) and this could also be the case, at least partly, for more major salinity changes recorded in the Purbeck sections. Difficulties are common in interpreting facies changes in such coastal settings, which were often freshwater and periodically cut off from the open sea. Climate, autocyclic processes, as well as tectonic events are also major control on facies distribution. Nevertheless, it is generally considered that the widespread Cinder Bed marked a sea-level rise at the middle-late Berriasian boundary, equivalent to a second-order MFS observed at the same time in other early Cretaceous European basins (MFS Be4/Be5, Jacquin et al., 1998). Following that scheme, inside the overall regressive trend, a transgression above the basal Dorset Purbeck unconformity allowed the infilling of basins, reaching its maximum during Cinder Bed deposition, followed by an overall regressive trend, as indicated by the growing freshwater influence at top of section, especially above the Corbula Member and until the Wealden facies (Fig. 2A).

### 2.4. Climate variation

Most authors concur that the Purbeck Limestone Group succession represents a major intensification of the seasonal semi-arid palaeoclimate that developed in the late Kimmeridgian–early Tithonian (Francis, 1984;

Fig. 2. Palaeoenvironmental succession of Durlston Bay and Lulworth Cove sections. A. Durlston Bay: synthesis after West (1975), Clements (1993), Westhead and Mather (1996), Batten (2002) and Radley (2002). Chronostratigraphy after Ogg et al. (1994), Feist et al. (1995), Abbink et al. (2001). Stratigraphical scheme, formations succession and informal members after Clements (1993), from base to top : “Cypris” Freestones Member, Hard Cockle Member, Soft Cockle Member, Marly Freshwater Beds, Cherty Freshwater Member, Cinder Bed, Intermarine Member, Scallop Member (S.M.), Corbula Member, Chief Beef Member, Broken Shell Member (B. Sh. Mbr.), Unio Member, Upper “Cypris” clay and Shale Member. Long-term sequence stratigraphy following the sequence-stratigraphy scheme at European scale of Jacquin et al. (1998), and highlighting the infilling of basal unconformity by shallow water deposits, the major marine invasion (Cinder Bed event) and the general regressive trend at top of section to the clastic Wealden facies beginning just above the Durlston Bay section. Palaeoclimatic interpretation showing the trend from semi-arid to semi-humid climates throughout the Berriasian after West (1975), Francis (1984), Deconinck (1986, 1993), summarized in Allen (1998), and this work. B. Lulworth Cove: long-term sequence stratigraphy and palaeoclimatic interpretation. Palaeoclimatic interpretation after West (1975) and Deconinck (1986), summarized in Allen (1998), and this work.

Wignall and Ruffell, 1990). The most successful documentation (to date) of the palaeoclimate change has been achieved through the use of plant fossils/microfossils (Francis, 1984) and clay mineralogy (Deconinck, 1986; Hallam et al., 1991; all data summarised in Allen, 1998). These studies suggest that the absence of hydrophytic pollen and kaolinite from the (lower) Lulworth Formation, which in addition contains gypsiferous beds with halite pseudomorphs, represents the extremity of the late Jurassic semi-arid palaeoclimate. The gradual increase in kaolinite contents through the (upper) Durlston Formation and into the clastic Wealden Group above, associated with the disappearance of evaporitic-bearing sediments, was taken by Allen (1998) to represent a gradual amelioration of the palaeoclimate to wetter conditions. At the same time, bathymetry probably did not change very significantly during deposition, and therefore cannot be used to explain the disappearance of sabkha facies in the upper part of Durlston Bay section.

A tectonic control on the kaolinite increase may also be invoked, when considering active extension, subsidence and uplifts occurring in the Wessex Basin at the Jurassic–Cretaceous boundary, as mentioned above. Nevertheless, such a control is unlikely, as:

- (1) there is no significant increase in illite amounts throughout the Lulworth and Durlston Formations, as it could be expected in case of uplift control on clay minerals inputs
- (2) the kaolinite increase in the Durlston Formation, as mentioned above, is a widespread pattern in the middle-late Berriasian of numerous outcrops, having been recorded in sections as far as in Tunisia (Schnyder et al., 2005).

A tectonic uplift could explain enhanced detrital inputs to basins, possibly associated with increasing hinterland soil erosion, but fails in explaining a widespread, synchronous change (using available chronostratigraphic data) of the nature of hinterland-derived clay minerals (kaolinite vs. illite and/or smectite), as it is observed at the Jurassic–Cretaceous boundary, on both margins of Tethys. A climate dominant control, suggesting a change in the nature of soils in the hinterland and an enhanced erosion, both associated with a more humid climate, is a much more powerful explanation.

In the Dorset area, such wetter conditions began just after deposition of the Cinder Bed and the related MFS (Deconinck, 1986, 1993), which correspond to the transition from Lulworth to Durlston Formation, well expressed at Durlston Bay section (Fig. 2A). At highest frequency, as pointed out above, salinity fluctuation recorded by molluscan and ostracoda faunas may be the result of climate-induced changes in runoff/evaporation balance (Radley, 2002; Horne, 2002).

### 3. SGR in palaeoclimate studies

#### 3.1. SGR-general

Gamma-ray and SGR logs are quick and commonly used methods of formation evaluation. Their use in sequence stratigraphy (van Wagoner et al., 1990), reservoir characterisation (Davies and Elliot, 1996), diagenesis, mineralogical determination (Hurst, 1990) and source-rock evaluation (Myers and Wignall, 1987) proves their wide application to sedimentology and stratigraphy. All of the above applications depend on mineralogical variation controlling the sites in which K, U and Th may reside. K is common in many sediments which bear feldspar, mica, clays or salts. U and Th have a number of host minerals in sedimentary rocks including clays, feldspars, heavy minerals, phosphates and organic matter. In addition, specific heavy minerals have high Th or U contents, creating a potential problem in interpreting the gamma-ray emission of sedimentary rocks containing zircons, uraninites or thorites (Hurst, 1990). Other problems in interpretation may be caused by K-rich feldspars in sandstones, organic matter content and anoxia (Myers and Wignall, 1987) and carbonate contents (McRoberts et al., 1998).

#### 3.2. SGR and palaeoclimate variation

K is known to be leached from feldspars and muscovite during kaolinite formation under conditions of hot and humid climates whilst Th is considered at least partially insoluble and concentrated during weathering (Parkinson, 1996; Osmond and Ivanovich, 1992). U is also considered more soluble than Th and thus prone to mobilisation during leaching and clay mineral diagenesis. Oxidizing conditions

in soils and rocks cause the U to take on the 6+ valence and form soluble complex ions. Conversely, reducing environments cause the U to take on the 4+ valence and becomes insoluble (Osmond and Ivanovich, 1992). Th/U ratios from shallow-buried mudstone-rich successions may show similarities to Th/K ratios, if the U is also being preferentially leached at source (Rosholt, 1992; Osmond and Ivanovich, 1992). Th/K ratios in mudrocks have been cited as a palaeoclimate indicator (Parkinson, 1996; Ruffell and Worden, 2000) where elevated Th contents (compared to K) may reflect a humid hinterland palaeoclimate in which K was leached from the clays; an hypothesis which has yet to be tested. We consider weathered mudstones as the most likely detrital materials to show this Th/K and Th/U separation as clay particles will have a relatively high surface area during weathering and impermeable properties when deposited.

## 4. Methods

### 4.1. Clay mineralogy

Clay mineral associations have been studied using X-ray diffraction (XRD) on oriented mounts. The samples were decarbonated, and then the clay fraction (<2  $\mu\text{m}$ ) was separated by sedimentation and centrifugation using the analytical procedure of Holtzapffel (1985). X-ray diffractograms were obtained using a Phillips PW diffractometer with  $\text{CuK}\alpha$  radiation and Ni filter. The identification of clay minerals was made according to the position of the (001) series of basal reflections of the three X-ray diagrams (Brown and Brindley, 1980; Moore and Reynolds, 1989). Semi-quantitative evaluations are based on the peak heights and areas summed to 100%, the relative error being 5% (Holtzapffel, 1985). The mean sample distance was 0.75 m at Durlston Bay (133 samples) and 0.5 m at Lulworth Cove (55 samples).

### 4.2. SGR

Our measurements were made using a Scintrex GIS-5 machine, calibrated to a Th standard and used on a 0.35 to 0.25 m sample distance, depend-

ing on lithology as pure carbonate beds were avoided. Only half our field SGR data set was used for bed-by-bed comparison with clay minerals, as the original SGR data set is more detailed than the clay minerals one. The detailed SGR data set shows that minor variations are observed in the SGR results at higher frequency, which confirms the general observed trends in SGR results. The methodology of Slatt et al. (1992) was followed, wherein five readings were taken at each site, the lowest and highest were discarded and the three remaining averaged. As a cross-check, further analyses have been undertaken using an Exploranium GR256 Spectrometer, allowing automatic derivation of Th/K and Th/U. The operational limitations and error margins of all these machines are described by Slatt et al. (1992) and Davies and Elliot (1996). Error margins are 5% for total counts, K and U, and 10% for Th.

### 4.3. Organic matter

Rock-Eval pyrolysis was completed using an Oil Show Analyser (OSA) machine (Espitalié et al., 1985a,b, 1986). The parameters presented here are the total organic carbon (TOC) content, in weight %, which reflects the quantity of organic matter in the sediment (carbon represents usually 50% to 70% of the total organic matter) and the  $T_{\text{max}}$  in  $^{\circ}\text{C}$ , used to estimate the degree of maturation of the organic carbon. The analyses were performed on the same samples than the clay mineralogy study.

## 5. Results

### 5.1. Durlston Bay (Fig. 3)

#### 5.1.1. Clay mineralogy

The clay fraction is composed of various amounts of chlorite (0% to 7%), illite (5 to 97%), illite/smectite mixed-layers (I/S, *smectites s.l.*) of  $R_0$  type (3% to 95%), kaolinite (0% to 30%) and the sporadic presence of palygorskite (0% to 10%). It has to be noted that palygorskite, in the small amounts observed (as at Lulworth Cove, see below) is difficult to estimate on X-ray diagrams and proportion presented here are indicative. The I/S mixed-layers predominate in the

clay fraction. Illite and kaolinite covary, in opposition with I/S mixed-layers.

Two mineralogical zones are distinguished at Durlston Bay:

- (1) Zone A is characterized by abundant smectitic minerals and by the absence of kaolinite, and occasional presence of palygorskite, until, at least, DB 91. The top of the zone is indicated by the first sporadic appearances of kaolinite in trace amounts (beds DB 112 topm.–DB 114), just above the Cinder Bed, at the base of the Intermarine Member. Two illite peaks (DB 100 and DB 104) are obvious below the Cinder Bed.
- (2) Zone B is characterized by both the appearance and subsequent increase in kaolinite content together with sporadic, continual traces of chlorite throughout. Above the first appearances of kaolinite (DB 112 topm.–DB 114), this mineral becomes a constant, but minor (less than 10%) part of the clay fraction above DB 124B–DB 130 base. The proportion of kaolinite increases to 10–15% above bed DB 134, until 20–30%. The rise of kaolinite begins just above the Cinder Bed, in several steps in the Intermarine Member. At the top of the section, there is some variations in the proportion of I/S mixed layers compared to kaolinite, with I/S mixed-layer clays increasing above DB 190, at the base of the Chief Beef Member.

In between the two main mineralogical zones, a smectitic rich interval can be observed between DB 68/74 and DB 134, with I/S mixed layers proportion generally above 65%.

#### 5.1.2. Spectral gamma-ray

Overall, total count gamma-ray radiation is controlled by K and U as all three display a similar pattern. Th is more scattered, suggesting that heavy minerals and carbonate have had no major effect on the data, otherwise spikes and cyclicity (respectively) would be apparent. Total counts fall from the base of the succession to the top, again matching the K and U contents. The total count curve, K and U present a clear stepwise fall pattern through-

out the section. K and U become erratic in the Mammal Bed to Cinder Bed interval, above DB 68–74. K shows a clear cyclic fall from the base to DB 100, a rise in DB 104 and then a fall. A marked drop is then observed in the K and U curve, above the Cinder Bed (around DB 112 topm.–114), some 10 m before the main influx of kaolinite (DB 130 base), where a new but smaller drop is observed in the total counts, K and U. Above DB 130–DB 145 (kaolinite peak), the total counts, K and U show a constant pattern, when at the same time, the kaolinite proportion reaches its maximum, above 10–15% (DB 134, see above). Th/K and Th/U show similar features to the overall counts, remaining low below the Cinder Bed, with a stepwise increase above. The DB 130 base limit is especially obvious, showing a sharp and marked increase in both Th/K and Th/U. Below the Cinder Bed, K and illite are correlated, above the Cinder Bed this relationship is less obvious. U is overall similar to K and thus illite.

#### 5.1.3. Organic matter measurements

In both Durlston Bay and Lulworth Cove sections,  $T_{\max}$  is mainly below 435/440 °C, thus indicating immature or slightly mature character of organic content. The main observation from the Durlston Bay section is the great heterogeneity in the organic matter distribution, with TOC varying from about 0% to the high value of 8.5% (DB 103). A 20 m-thick organic matter-enriched interval is obvious in the middle part of the section, between DB 88 (TOC=1.5%, and DB 130 top (TOC 2.8%). The organic matter-enriched interval comprises the Marly Freshwater Beds, the whole Cherty Freshwater Member, the Cinder Bed and the main part of the Intermarine Member. It is associated with mainly freshwater, lacustrine environments, as indicated by charophytes distribution (Fig. 3). Outside that specific 20-m thick interval in the middle part of the section, TOC values are generally under 2%, and mostly under 1%.

### 5.2. Lulworth Cove (Fig. 4)

#### 5.2.1. Clay mineralogy

The clay mineral suite at Lulworth Cove is similar to Durlston Bay, with illite (5% to 58%), I/S mixed

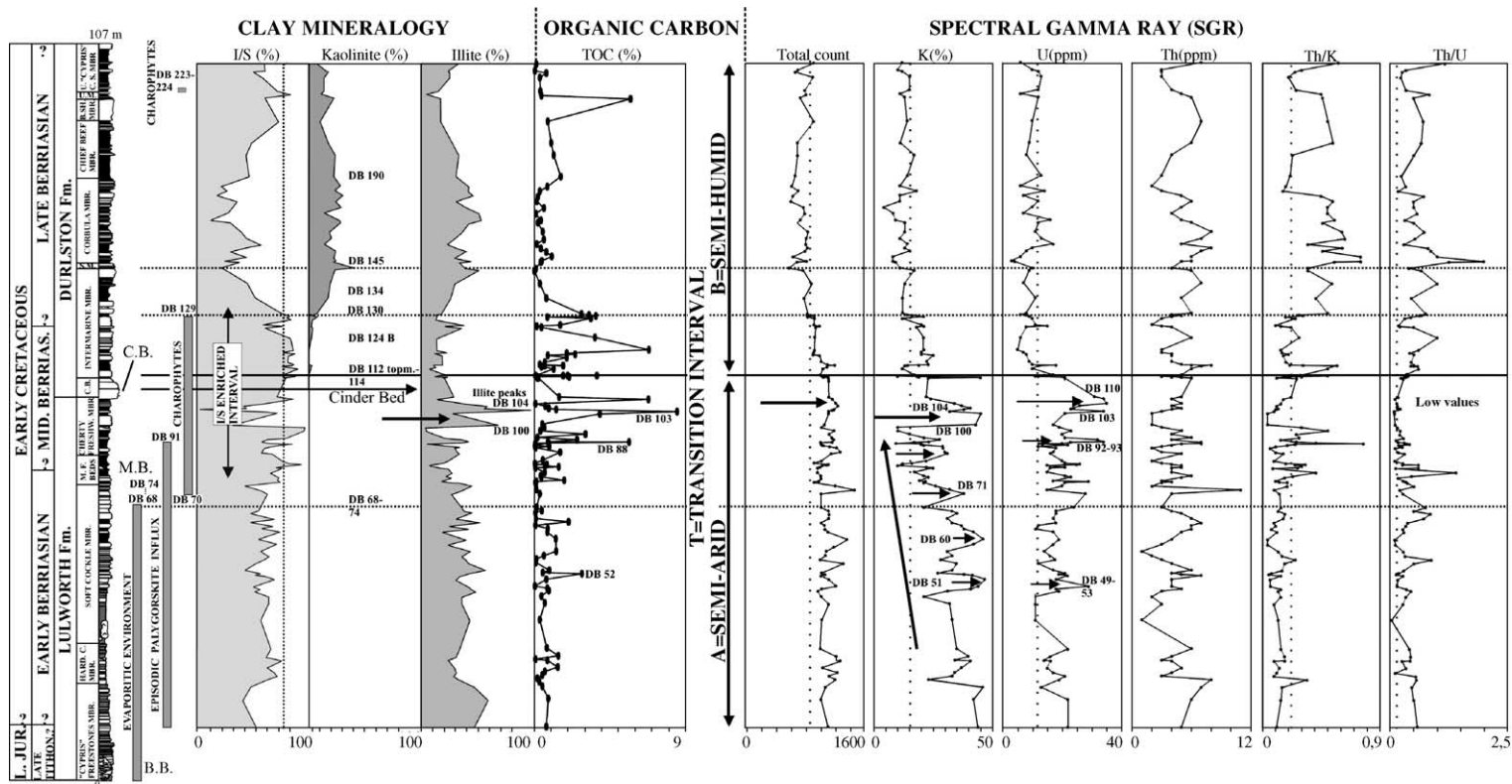


Fig. 3. Clay mineralogy, TOC (Rock-Eval), and SGR data of Durlston Bay section. Trace occurrences of chlorite and palygorskite are not shown on the figure. Vertical episodic distribution of palygorskite, having a paleoclimating significance, is indicated. Studied samples cited in the text are figured, with their bed number after Clements (1993), allowing precise position in his detailed log. Vertical distribution of main evaporitic facies after West (1975) and Clements (1993). Vertical distribution of charophytes after Clements (1993) and Feist et al. (1995). Horizontal continuous and dotted lines highlight respectively major and minor climate boundaries, showing a stepwise, three-part climatic trend (A = semi-arid, T = transitional, organic-rich interval, B = semi-humid). Observe the climatic-controlled rise of Th/K and Th/U in the SGR data.

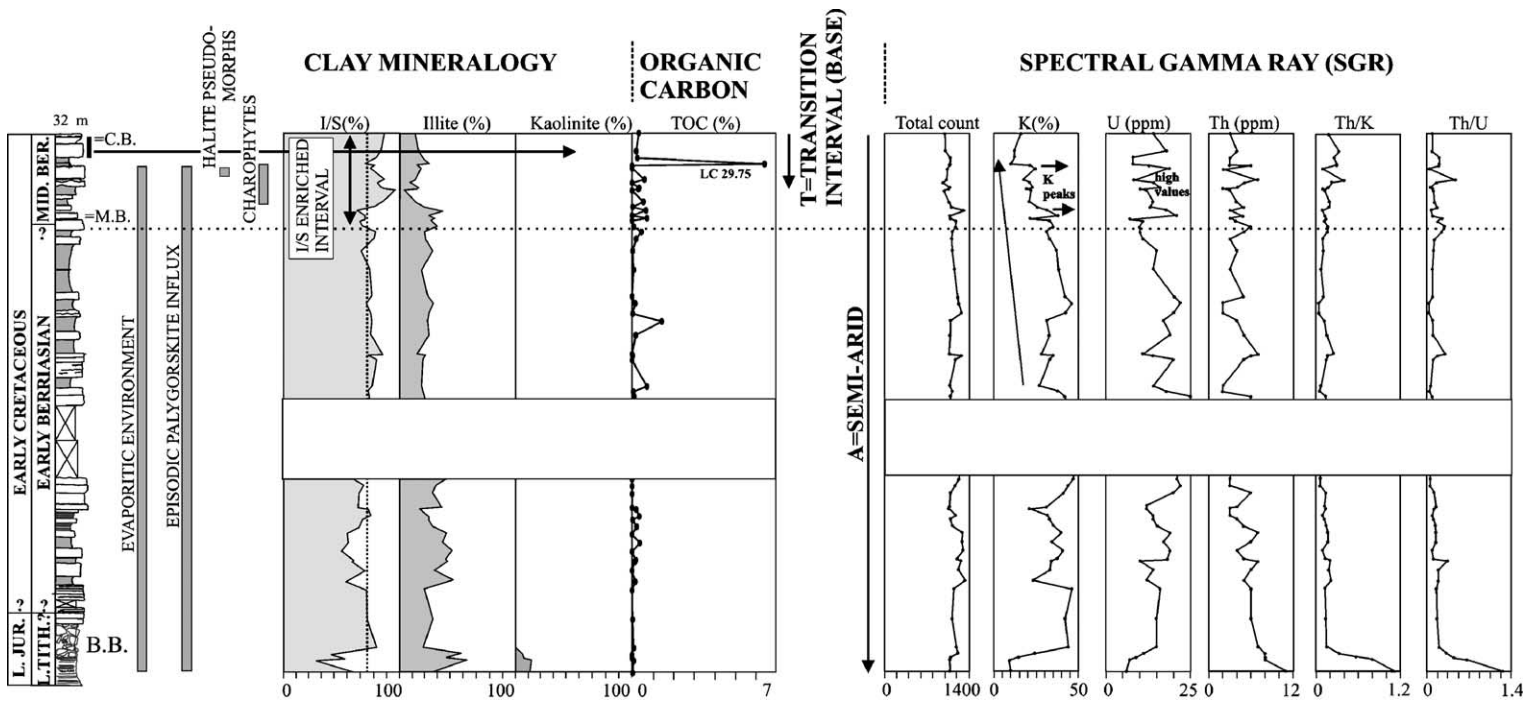


Fig. 4. Clay mineralogy, TOC (Rock-Eval), and SGR data of Lulworth Cove section. Traces occurrences of chlorite and palygorskite are not shown. Vertical distribution of palygorskite is indicated. Halite pseudomorphs distribution after Radley, 1992, and this work. Charophytes distribution, also this work. Horizontal dotted line highlight the first step of the climatic change, from semi-arid interval A to the transitional interval (compare to Fig. 3). Observe the beginning of the climate-controlled rise of Th/K (also compare to Fig. 3). Beginning of Th/U rise is less clear.

layers of R<sub>0</sub> type (28 to 96%), kaolinite present in small amounts (6 to 14%) (only at the base of the section), and palygorskite, again in small and sporadic amounts through the section. A difference with Durlston Bay is the absence of chlorite. As observed at Durlston Bay, I/S mixed-layers predominate the clay mineral fraction. The same zone A with absence of kaolinite and sporadic presence of palygorskite can also be recognized, if the small occurrence at base of the Lulworth Cove section is not taken into account. This zone is in the same stratigraphic position at Lulworth Cove, beneath the Cinder Bed. As at Durlston Bay, an intermediate smectitic interval is observed at the top of the outcrop, with I/S mixed layers proportions above 80%, and reaching a maximum of 96%, between the Mammal Bed and the Cinder Bed.

#### 5.2.2. Spectral gamma-ray

The overall pattern of spectral gamma-ray response from the Broken Beds to the Cinder Bed seen at Durlston Bay is observed at Lulworth Cove, thus showing a lateral continuity of data. The start of the fall in total counts, K and U is observed, although the section from Cinder Bed upward, where there were remarkable changes at Durlston Bay, are not preserved at Lulworth Cove. K and U become erratic between the Mammal and the Cinder Bed, again as observed at Durlston Bay. A minor complication is the presence of low K and U values, and therefore high Th/K and Th/U at the base of the section under the Broken Beds, together with small amount of kaolinite in the clay mineral assemblages.

#### 5.2.3. Organic matter measurements

The organic matter is generally less well-preserved at Lulworth Cove than at Durlston Bay, with TOC values being under 1% along most of the section. The top of the section shows an enrichment in organic carbon in the last two meters (sample LC 29.75: TOC=6.45). This level, located just under the Cinder Bed, is a lateral equivalent of the base of the organic matter-enriched interval at Durlston Bay. It is also associated at Lulworth Cove with charophytes occurrences (Fig. 4), showing at least a 20 km lateral extension for the organic-matter enriched lacustrine environments.

## 6. Discussion

### 6.1. Clay mineralogy and palaeoclimate

#### 6.1.1. A three stage palaeoclimatic evolution

The high resolution clay mineralogy data set at Durlston Bay and Lulworth Cove sections presented here allows us to refine the previously presented palaeoclimatic evolution. Comparison with available sedimentological data show a remarkable three parts stepwise palaeoclimatic change, from semi-arid below the Cinder Bed (early Berriasian) to semi-humid climate above (middle-late Berriasian) (Figs. 3 and 4):

- (1) The semi-arid zone A is characterized by presence of evaporites, absence of kaolinite, episodic presence of small amount of palygorskite. Zone A extends from base to DB 112 topm.–DB 114. The environment is considered as evaporitic lagoons and sabkha facies during deposition of the Cypris Freestones Member, the Hard Cockle Member and the Soft Cockle Member, passing to mostly freshwater to locally hypersaline facies above.
- (2) Above DB 112 topm.–DB 114, the semi-humid zone B comprises a stepwise development of a more humid phase, indicated by the rise of kaolinite. It comprises the Intermarine Member, the Scallop Member, the Corbula Member, the Chief Beef Member, the Broken Shell Member, the Unio Member, and finally the Upper Cypris Clays and Shales Member. Above the marine incursion of the Cinder Bed, the environment is mostly freshwater, fluvial to lacustrine and brackish water dominated, with minor marine incursions (e.g. the Scallop Member), as indicated by the molluscan and ostracoda faunas. The freshwater influence become clearly predominant at the top of the section, above the Corbula Member, where connections with the shallow sea seem to disappear.
- (3) In between the two previous zones, a transitional interval (T) is recorded, starting around DB 68–74 and ending at DB 130 base, characterized by the absence of evaporites, absence of palygorskite and continued absence of kaolinite. This zone comprises the Marly Freshwater Beds, the Cherty Freshwater Bed, the Cinder Bed marine incursion, and the main part of the

Intermarine Member. This correspond to a mostly lacustrine environment, as indicated by charophytes distribution, which was, at least episodically, connected with the marine realm.

The Lulworth Cove section is a lateral equivalent of the base of Durlston Bay section and shows clearly the same trends: semi-arid phase, and then beginning of the transitional phase until the Cinder Bed.

#### 6.1.2. *Dry/wet cycles in the transitional interval?*

The transitional climatic system is also marked by highest I/S mixed-layers proportions, both in Durlston Bay (between DB 68/74 and DB 130 base) and Lulworth Cove (at the top of the section) (Figs. 3 and 4). This could be partly a result of formation of authigenic Mg-rich smectitic minerals. Such minerals formation is known in ancient examples in lakes or marine coastal shallow settings, in palaeoenvironments submitted to high evaporation rate, and chemical confinement, with concentration of saline and/or alkaline solutions (Chamley, 1989). A minor authigenic formation of smectites in the lacustrine and saline environment of the Purbeck succession in Dorset, together with the dominant input of detrital smectite from inland erosion, could have led to highest proportion of smectitic minerals in the lacustrine organic matter-rich transition interval. Illitic-rich clay fraction, probably formed at surface temperature by authigenic processes, have also been described in Purbeckian facies of the French and Swiss Jura (Deconinck et al., 1988) and in the Dorset area, at Durlston Bay (Deconinck, 1986). Such illite formation has also been proposed for the Jurassic, alkaline, saline lake T'oo'dichi in Colorado (Turner and Fishman, 1991). Illite is thought to have developed by illitization of smectite in a potassic alkaline solution, during successive cycles of wetting and drying, marked by evidence of subaerial exposure (Deconinck et al., 1988, 2001). The K<sup>+</sup> ions necessary for illite formation by illitization of smectite may be supplied by various sources (e.g. erosion of volcanic rocks) but an input of K<sup>+</sup> by marine waters during ingressions of "Purbeckian" coastal shallow-water environments was favoured by Deconinck et al. (1988). The illitic peaks DB 104 (up to 97%), and possibly DB 100 (around 68% of the clay fraction), in the Cherty Freshwater Member at Durlston Bay (Fig. 3), as pointed by Deconinck

(1986), may reflect, together with evidence of subaerial exposure indicated by numerous mudcracks in the interval, alternation of dry/wet cycles and episodically occurrence of high evaporation rate and partly chemical confinement. These data suggest the continuation of episodic and/or seasonal dryness, despite the stepwise rise of humidity in that transitional palaeoclimatic phase. An alternative hypothesis for high I/S proportions could be a smectite authigenesis associated with condensation in low accumulation rate settings, as it could have been the case when considering organic matter accumulation during the transitional interval. Nevertheless, occurrence of small pseudomorphs after halite in some beds of the Cherty Freshwater Member and in the cherty equivalent beds at Lulworth Cove (Radley, 1992, and this work, Fig. 4) better support the hypothesis of dry/wet cycles.

The clay mineralogy data set thus details more precisely than previously published works the paleoclimate evolution at the Jurassic–Cretaceous boundary in the Dorset, showing a clear, stepwise palaeoclimatic evolution which can now be compared with SGR data.

## 6.2. *SGR data and palaeoclimate*

### 6.2.1. *General trend*

The SGR data underline the major environmental and palaeoclimatic evolution suggested by clay mineralogy, both in Durlston Bay and Lulworth Cove sections (Figs. 3 and 4):

- (1) the overall total count, K, and U fall and the subsequent Th/K and Th/U rise from base to top of sections follow the rise in kaolinite and the change from evaporitic, semi-arid, to more humid environment.
- (2) the erratic behaviour of K and U between the Mammal Bed and the Cinder Bed begins just above DB 68–74 at Durlston Bay, in the Marly Freshwater Beds, as the proposed transitional climatic system occurs, as indicated by clay mineralogy, and when the main sabkha facies of the semi-arid phase are disappearing. The total count is quite stable between the Mammal Bed and the Cinder Bed because as K values are mostly falling, U values rather increased.
- (3) above, the stepwise fall of K, U and subsequent rise of Th/K and Th/U show three major bound-

aries. A first marked drop in K, U, total count above the Cinder Bed (DB 112 topm.–DB 114) coincides with the first occurrence of kaolinite. The new but smaller drop at the base of DB 130 is symmetrical with the main influx of kaolinite, and the end of the transitional, enriched I/S interval, suggesting a new step in the increasing atmospheric moisture. Finally, the rather low and constant K, U, total count values, and high Th/K and Th/U values above DB 145 characterize the maximum atmospheric moisture.

The Lulworth Cove section also shows the beginning of the general drop in K, U, and total count values (Fig. 4), especially in the top of the section when successively lateral equivalent of the Durlston Bay's Mammal Bed, Cherty Freshwater Beds and Cinder Bed were deposited.

A stepwise fall of K, U and total count values and correlative rise of Th/K and Th/U values from base to top of sections is the major trend on SGR data, and is parallel to kaolinite rise in clay mineralogy. This could be interpreted as the result of the general sea-level fall recorded by facies succession from the basal Purbeck unconformity to the Wealden sandy facies, which begins just above the Durlston Bay section studied. Sand/silty-enriched carbonate and shaly beds are quite common in the second-half of the Durlston Bay section (Clements, 1993), associated with more proximal/fluvial facies. Such a sea-level controlled coarsening-up and quartz-enriching pattern relative to clay could lead to lower K, Th, and U values, these elements being sited in clays, and correlative higher Th/K values (Davies and Elliot, 1996; Parkinson, 1996). Nevertheless, we interpret this general pattern as being mostly control by the climate evolution to wetter climate, the SGR data underlining accurately the stepwise climatic boundaries previously suggested by clay mineralogy and facies succession. The fall in K begins at Durlston Bay section above DB 51–DB 60 in the Soft Cockle Member, well before any sea-level controlled sand/silt enrichment, which occur at top of section. A palaeoclimate change from the semi-arid interval A to the more humid, transitional interval (T) may help explain this pattern.

We thus propose that the concentration of Th during weathering and palaeoweathering (Rosholt, 1992; Osmond and Ivanovich, 1992) favours the link

between hot and humid conditions indicated by the rise in kaolinite, mobilising and leaching K and U whilst concentrating Th in the hinterland soils. In such a scenario, detrital clays, relatively depleted in K and U are trapped in the basin. The clays in the Dorset being mainly detrital ones (apart from some possible minor authigenic clays occurring in the transitional interval), we can then suggest that detrital clays trapped in the basins, relatively depleted in K and U and enriched in Th, due to the climate variation, composed the main part of the SGR signal. The low total counts values above the Cinder Bed are in such a scenario also associated with enhanced continental leaching, reducing the overall SGR signal in soils, and then in detrital exported clays.

However, a complication could arise when considering that U can go in solution in stream waters, after having being leached in hinterland soils. Such U in solution could then be bound in the basin with organic matter in association with reducing, anoxic/dysoxic conditions and/or be bound with phosphates, which could complicate the final SGR record (e.g. Schmoker and Hester, 1983; Osmond and Ivanovich, 1992; Parkinson, 1996). In addition, heavy minerals, enriched in U, could also be trapped in the basins, associated with detrital pulses. These possible problems can be resolved when considering that organic matter is generally present in minor amounts, apart from the organic-rich transitional interval and that the possible occurrence of U bound to organic matter (in the middle part of Durlston outcrop and at the top of Lulworth Cove outcrop) cannot explain the main trends of the SGR curves, which are characterized by clear, continuing trends from base to top of the outcrops. Possible relationships between organic matter and SGR will be discussed in the next section. In addition, in the Dorset examples, we measured mainly mudstones, with no widespread phosphate deposits, and with few sandstones and thus few heavy minerals. We thus consider here the climate change to be the major control on SGR signal.

Th/K and Th/U ratios show the same increasing trend throughout the sections, suggesting that both ratios have a palaeoclimatic value. However, as it could be hypothesized, the Th/U rise is less marked than the Th/K, and thus the Th/K curve has a better sensitivity to changes in moisture conditions. This may suggest that SGR data could have, to some extent

and at least in these sections, a greater sensitivity to rising moisture than clay mineralogy, because falls in K, U values and correlative rise in Th/K ratios begin above the Mammal Bed in the climatic transitional interval (T), before the rise in kaolinite. It could be possible that the inputs of kaolinite from the weathering profiles could be marked by a threshold effect in moisture—a certain moisture value being necessary for the clay mineralogical soils nature to change in hinterland. A time-lag would then have existed between moisture changes and kaolinite inputs to basins, as suggested by Thiry (2000). The observed discrepancy with Th/K ratios would perhaps suggest that the kinetic of K leaching and Th concentration in soils could be different, faster than kinetic of kaolinite formation, and thus with a better response to palaeoclimate change: this requires further examination.

Only a few works have focussed on the usefulness of SGR data as climatic tool and SGR/clay mineralogy relationships (Hesselbo, 1996; Ruffell and Worden, 2000; Deconinck et al., 2003), and this study adds high resolution data on a well-documented climate change. Nevertheless, caution must be taken as Th/K and Th/U ratios, which in such interpretation reflect more or less the kaolinite content of the clay assemblage, do not automatically reflect a moisture curve. As an example, in the Cenozoic

study of Deconinck and Vanderaverroet (1996) and Hesselbo (1996), Th/K and Th/U ratios are correlated to kaolinite variations. Nevertheless, the observed kaolinite rise, correlated to illite variation, is interpreted as a strong intensification of hinterland erosion linked to sea-level lowering and global cooling, rather than only a rise in moisture.

#### 6.2.2. SGR and organic matter

There is no direct correlation between U and organic matter both in Durlston Bay and Lulworth Cove (Fig. 5), but the climatic imprint on the SGR signal is however sometimes modulated by superimposed effect of organic content.

Highest U, variable values between DB 68 and the base of the Cinder Bed at Durlston Bay and the same trend observed at the top of the Lulworth Cove section between the lateral equivalent of the Mammal Bed and the Cinder Bed has to be noted, as it correspond to the organic matter-enriched interval in both sections. It is significant, as at the same interval, Th values are stable but variable, and K values show a clear decreasing trend, which could be considered as climate-controlled (Figs. 3 and 4). U peaks are noted in the Durlston Bay section, on DB 92 mid.– 93 mid. and DB 103–110 top interval in the Cherty Freshwater Member, associated with a high organic matter con-

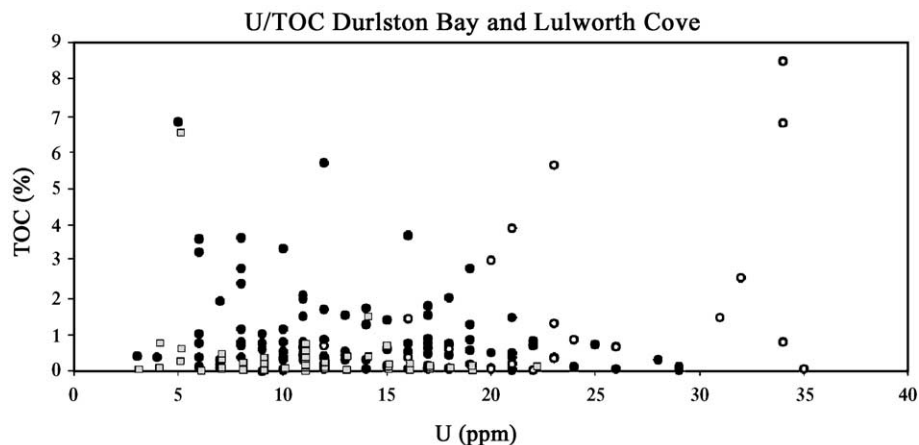


Fig. 5. Uranium (SGR data) versus TOC diagrams of Durlston Bay (black and white circles) and Lulworth Cove (grey squares) sections. No direct correlation is observed between U and TOC in both sections, showing that organic matter is not the major control on uranium concentration throughout the sections. Nevertheless, the samples from the organic-rich interval at Durlston Bay (white circles) are mostly in the high U-values part of the diagram, showing that there is an influence of organic matter on uranium concentration, at least in this interval (also see Fig. 3). This pattern is not observed at Lulworth Cove, but this section only show a part of the organic-rich interval, and one can observe a positive shift and quite high U values when entering in this organic-rich interval at top of section (Fig. 4).

tent (respectively, TOC=0.8–2.55% and TOC until 6.7–8.5%). U peaks are also recorded on DB 49–53 beds, in the Soft Cockle Member (under the organic matter-enriched interval for the latter samples). The DB 49–53 U-rich beds, even out of the main organic matter-enriched zone, correspond to the first organic-rich samples from the base of the Durlston Bay section (DB 52, TOC=2.8%, Fig. 3), which resemble that of the organic matter-enriched interval. This pattern probably reflects the influence of the high organic content of the organic matter-enriched interval on the U concentration, high uranium concentration being often associated with organic matter (e.g. Schmoker and Hester, 1983; Osmond and Ivanovich, 1992; Parkinson, 1996). Uranium can be directly adsorbed onto organic matter, and/or can be precipitated, taking on its 4+ valence under reducing, anoxic conditions associated with organic matter preservation (Davies and Elliot, 1996; Parkinson, 1996). The high U values and peaks could then reflect the anoxic/oxic state of water masses, indicating, at least partly, anoxic conditions between the Mammal Bed and the Cinder Bed. In the particular zone between Mammal Bed and Cinder Bed, the U curve could also partly reflect condensation associated with a low sedimentation rate, in time of rising sea-level. The transgressive surface of the Cinder Bed is indicated by a U peak, low Th/K, and a peak in the total count, just below the Cinder Bed, between DB 104 and DB 112, a characteristic pattern of such surface (Davies and Elliot, 1996).

However, at Durlston Bay, U values drop above the Cinder Bed, whereas the TOC values remain high until DB 130 base, in the Intermarine Member. This discrepancy between U and TOC content could correspond to a change in the dominant control upon the U concentration above the Cinder Bed, when humid conditions are rising. Fall of K and U values due to leaching from soils in the hinterland now dominate the SGR distribution, and overcome the influence of organic matter concentration (organic matter leading to higher U values), whereas the organic content remains high. If correct, this would be an example of the contrasting and evolving controls on SGR signal.

### 6.2.3. Dry/wet cycles and K values

Illite peaks in the clay assemblage can also be compared to the SGR signal. The illite peaks of DB 100–104 in the Cherty Freshwater Member at Durl-

ston Bay correspond to K peaks in the SGR data (Fig. 3), illite being an K-rich clay. As suggested above, these illitic-rich levels may correspond to illitization of smectites at surface temperatures during successive dry/wet cycles in the transitional palaeoclimatic interval. Therefore, the K curve could in such peculiar circumstances reflect dry/wet cycles. Other K peaks are obvious in the semi-arid zone A and the organic-rich transitional interval T (Fig. 3): K peaks centered on DB 51A, DB 60 and DB 71. The concentration of such K peaks and high variability of K values in the first half, more or less dry part of the Durlston Bay section, whereas such peaks are missing in the second half, more humid zone of the section, suggest a climatic control on that pattern, as hypothesized. This pattern is less obvious at Lulworth Cove section, but K peaks also occur (Fig. 4).

## 7. Conclusions

A high resolution SGR study, combined with clay mineralogy and organic carbon analysis has been performed on two sections of the coastal shallow water, marine and non-marine facies of Dorset, southern U.K. The focus of the work was to test the sensitivity of SGR to climate, on a previously well-documented climate change, as few previous papers having theorized the climatic control on SGR data.

The conjunctive use of data show a three-stage climatic evolution throughout the Berriasian, a middle Berriasian transitional climatic phase being sandwiched between a late Tithonian–early Berriasian semi-arid phase and a late Berriasian semi-humid phase. The transitional climatic interval is associated in mostly lacustrine settings with persistence of dry/wet cycles.

The SGR data show a general fall of total count, K, U values, and a subsequent rise of Th/K and Th/U ratios throughout the Berriasian. This pattern follows other climatic-related proxies (e.g. clay mineralogy, evaporites distribution) and is interpreted as having a strong palaeoclimate control. Comparison between the various data suggest that SGR can be a monitor of moisture change. This study confirms the power of SGR as a quick and cheap reconnaissance tool in outcrop palaeoclimatic studies as underlined by Ruffell and Worden (2000), but recommends that as many

conjunctive analyses as possible are used in development of integrated palaeoclimate-sea-level-tectonism models. When palaeoclimate is suggested to influence the SGR data (especially, it seems the Th/K ratio), only local hinterland weathering or sediment source can be suggested until a widespread, possibly palaeoclimatic origin can be established. To achieve this aim, the best use of SGR data as palaeoclimatic tool is probably:

- 1) to properly identified and characterized the climatic signal by an integrated study in few reference sections, using in parallel SGR and other climatic proxies
- 2) once the SGR signal has been “calibrated”, to produce an extensive analysis of numerous outcrops and/or boreholes by SGR, this method being quick and cheap. In addition, phosphate, organic matter-rich deposits and heavy minerals associated with sand levels must be avoided or accounted for.

By contrast, when using Th/K and Th/U ratios for other purposes, such as identifying episodes of anoxicity in source-rocks or flooding surfaces in reservoir seals, the possibility of a palaeoclimate control on SGR data must also be considered and discussed to avoid misinterpretation.

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