

# Environmental and climatic controls of the clay mineralogy of Albian deposits in the Paris and Vocontian basins (France).

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1	Environmental and climatic controls of the clay mineralogy of Albian deposits in
2	the Paris and Vocontian basins (France)
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L4	
<b>L</b> 5	Abstract
L6	High-resolution clay mineral analyses were performed on lower and middle Albian
L7	deposits from the Paris and Vocontian basins in order to specify the weathering conditions
L8	that prevailed at that time. The clay mineral assemblages are composed of small proportions
L9	of chlorite and vermiculitic clays associated with abundant illite, R0 type illite-smectite
20	mixed-layers (smectite) and kaolinite. Clay minerals originated from the physical alteration
21	and chemical weathering of rocks and soils outcropping on the Variscan massifs bordering the
22	studied areas. In the Paris Basin, the covariation of illite and kaolinite suggests the reworking
23	of these latter minerals from palaeoalterations and saprolites coupled with an intense runoff
24	on the London-Brabant Massif under a warm-temperate climate.
25	In comparison with the Paris Basin, the lower Albian series deposited in the Vocontian
26	Basin are enriched in smectite because of the preferential deposition of this mineral in deep
27	offshore environments. In this basin, illite and kaolinite did not show any covariation
28	suggesting that kaolinite is rather the chemical weathering product of rocks exposed in the

French Massif Central. The stratigraphic distribution of clay minerals in both studied areas is

consistent with a cooler and dryer climate around the Aptian/Albian transition. An enhanced

precipitation rate is suggested by an increasing proportion of kaolinite from the

Douvilleiceras mammillatum Zone upward. This humid interval ended in the Anahoplites

intermedius Zone contemporaneous with the smectite-rich sedimentation in the Paris Basin

revealing semi-arid conditions consistent with palynological data. This semi-arid interval

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encompassing the *Dimorphoplites niobe* and *D. biplicatus* zones (and possibly the *Dipoloceras cristatum* Zone) is interrupted at the base of the *Mortoniceras pricei* Zone by a return to a more humid climate.

Keywords: Clay minerals, Albian, Paris Basin, Vocontian Basin, Palaeoclimate

# 1. Introduction

Most often, the reconstruction of Cretaceous climates is based on an estimate of the ocean surface or deep water temperatures from a variety of proxies including  $\delta^{18}$ O, TEX<sub>86</sub> and more recently  $\Delta_{47}$  (Friedrich et al., 2012, O'Brien et al., 2017; Meyer et al., 2018). The global climate evolution during the Albian is well known in terms of temperatures inferred from numerous  $\delta^{18}$ O data measured on foraminifera and belemnite guards (Friedrich et al., 2012). After the late Aptian 'coldhouse' climate (Herrle and Mutterlose, 2003; Pirrie et al., 2004; Bodin et al., 2015; Rodríguez-López et al., 2016), the Albian stage is characterized by a regular increase in temperature from the base to the top. This trend continued until the Cretaceous thermal maximum that occurred around the Cenomanian/Turonian boundary (O'Brien et al., 2017; Huber et al., 2018).

The estimation of the fluctuations in the hydrological cycle and the intensity of the precipitation during the Albian is discussed much less often (Föllmi, 2012), even though this stratigraphic interval corresponds to dramatic changes in the Earth's history. Albian environments are notably characterized by a radiation of angiosperms and their development in floodplain areas and by the emergence of a narrow humid equatorial climatic belt in comparison with the earlier stages of the Early Cretaceous during which the equatorial humid belt was absent (Chumakov et al., 1995; Poulsen et al., 2001; Chaboureau et al., 2014). A warm and humid belt around the equator is deduced from the common occurrence of bauxite and coal during the Albian (Chumakov et al., 1995). This re-organization of the climatic belts is likely a consequence of the widening of the South and North Atlantic Oceans which thereby introduced a source of moisture (Chaboureau et al., 2014). Therefore, it appears that the Albian was a pivotal period from a climatic point of view that may have had consequences on the hydrological cycle.

In the Anglo-Paris Basin, Albian seawater temperatures were estimated using  $\delta^{18}O$  analysis of the belemnite guards and fish teeth from the Wissant section outcropping in the Boulonnais area (northwest of the Paris Basin, Bomou et al., 2016, Fig. 1). The measured

temperatures on the belemnite guards (bottom waters) range from 13.5 to 19.3°C with an increase from the middle to late Albian. The  $\delta^{18}$ O values for the phosphate in the fish teeth indicate temperatures (from intermediate or sea surface waters) that varied between 22° and 28°C with a similar trend. Increasing temperatures during the Albian are consistent with the long-term global warming recorded from the Albian to the Cenomanian using either  $\delta^{18}$ O measured in the foraminiferal test or the TEX<sub>86</sub> proxy (Friedrich et al., 2012; O'Brien et al., 2017; Kanungo et al., 2018). However, although moisture is a key palaeoclimate parameter, the changes in humidity and weathering conditions that occurred during the Albian are much less documented (Ufnar et al., 2002; Herrle et al., 2003b; Horikx et al., 2017). Our objective is therefore to improve knowledge on humidity/aridity conditions prevailing during the Albian using clay mineral assemblages from sediments deposited in two distinct provinces: the Paris basin (Boreal Province) and the Vocontian basins (Tethyan Province).

# 2. Geological context

During the Albian, the Anglo-Paris Basin was an epicontinental sea connecting the Boreal domain to the north-west and the Tethyan domain to the south-east. It was located at a latitude close to 35°N within a mid-latitude warm humid belt (Fig. 1, Boucot et al., 2013; Amédro and Matrion, 2014; Gély et al., 2014). The Albian deposits from the type locality (stratotype, department of Aube, south-east of the Paris Basin) were only known by temporary outcrops or quarries where only a few metres of the sedimentary succession can be observed (Amédro, 1992; Amédro and Matrion, 2007; 2008, 2014). Consequently, a complete continuous succession could not be established in this part of the Paris Basin. Recently, boreholes were drilled by ANDRA (French agency for nuclear waste disposal) in the department of Aube, allowing the whole lower to middle Albian clayey succession to be examined. The sedimentary series belongs to the so-called 'Argiles tégulines de Courcelles' unit which overlies the Aptian and lowermost Albian greensands, the so-called 'Sables Verts de l'Aube'. In one of the boreholes, the succession encompasses the lowermost part of the overlying 'Marnes de Brienne' of late Albian age.

The Paris Basin was bordered to the south-west by the Armorican Massif and the French Massif Central, and to the north-east by the London-Brabant Massif (Fig. 1, Amédro and Matrion, 2014; Amédro and Robaszynski, 2014). At that time, north-west Europe was made up of large islands inherited from the Variscan orogeny. Although clay minerals are likely to be deposited very far from their detrital source, the French Massif Central and the London-Brabant Massif were probably the main sources of detrital clay mineral since coeval

detrital sandy deposits of the Paris Basin originated from these massifs (Amédro and Robaszynski 2014). These massifs were located at slightly different latitudes and are made up of distinct parent rocks, therefore, significant differences in the composition of clay assemblages can be expected.

The Vocontian Basin was located on the northern margin of the Tethys Ocean, around 30°N at a somewhat lower palaeoaltitude and closer to the semi-arid belt than the Paris Basin (Chumakov et al., 1995, Hay and Floegel, 2012; Fig. 1). This basin was surrounded by shallow platforms except to the east where it was largely opened to the Tethys Ocean (Fig. 1). Lower to middle Albian deposits (Blue Marls Formation) are well-exposed in several sections (Bréhéret, 1997; Gale et al., 2011). The Col de Pré-Guittard section was chosen for this study considering its location in the western part of the Vocontian Basin, far from thermal influences that would transform detrital clay minerals. This reference section is also clearly defined with biostratigraphy (Kennedy et al., 2000, 2014; Petrizzo et al., 2012), and was approved recently as the Albian Global Boundary Stratotype Section and Point (GSSP) (Kennedy et al., 2017).

# 3. Lithology and biostratigraphy

#### 3.1. Boreholes from the Paris Basin

Two boreholes (AUB111 and AUB220) located close to Juzanvigny, and one borehole drilled close to Vallentigny (AUB240) were studied (Fig. 2). Samples were taken every meter in the borehole AUB240 and every 50 centimeters in the boreholes AUB111 and AUB220.

The distance between boreholes AUB111 and AUB220 is only 1 km. One of the cores (AUB111, Fig. 3) has been described in detail including a precise biostratigraphic scheme on the basis of the abundant ammonite fauna (Amédro et al., 2017) following the ammonite biozonation proposed by Amédro and Matrion (2014). Lower to middle Albian deposits (Argiles tégulines de Courcelles) essentially consist of bioturbated clays and silty clays with occasional pyrite and phosphate nodules (Knight 1999). Phosphate nodules are either scattered or in the form of thin horizons that may be correlated at a regional scale. The base of the unit is more or less sandy, with increasingly more abundant glauconite further down the core, until the boundary with the Aptian greensands (Sables Verts de l'Aube Formation). Borehole AUB 220 shows a sedimentary succession quite similar to that described in borehole AUB111, but it can be used to provide more descriptions further upward in the series (Figs. 4 and 5; Supplementary Appendix). In addition, the recovery across the boundary between the Sables Verts de l'Aube and the Argiles tégulines de Courcelles is better than in the AUB111

borehole. Notably, a condensed horizon composed of a 10 cm-thick phosphatic crust corresponding to a hardground is particularly well preserved in this borehole (Fig. 5). The age of the deposits recovered in the lower part of the two boreholes is not well established. In this part of the Paris Basin, the Sables Verts de l'Aube unit is considered to be Aptian in age (Amédro and Matrion 2007) because of the occurrence of the benthic foraminifera *Verneuilinoides subfiliformis* which suggests that the Sables verts de l'Aube belong to the *Hypacanthoplites jacobi* Zone from the upper Aptian (Damotte & Magniez-Jannin, 1973; Mutterlose et al., 2003).

The phosphatic crust corresponds to a condensed interval that, according to previous studies on outcrops located in the same region, likely includes the *Leymeriella tardefurcata* and *Leymeriella regularis* ammonite zones (Amédro and Matrion 2007, 2014). At the base of the two boreholes, the 5 m-thick interval between the phosphatic crust and the first occurrence of *D. mammillatum* is not well dated as only *Beudanticeras* sp. have been found. This interval probably successively corresponds to the *Sonneratia kitchini, Cleoniceras floridum* and *Hemisonneratia puzosiana* zones compared with the sections outcropping to the south-east of the Paris Basin (Destombes *in* Rat et al., 1979; Amédro et al., 2000; Amédro and Matrion, 2008). Above this poorly dated interval, the borehole is precisely dated by numerous ammonites which can be used to draw an accurate biostratigraphic scheme (Figs. 3-5). The two boreholes can be easily correlated in particular by using marker beds that include horizons of phosphate nodules and bio-events (Amédro et al., 2019).

The change from the Aptian greensands, deposited in shoreface, estuarine or fluvio-deltaic environments (Guillocheau et al., 2000), to the Argiles tégulines de Courcelles deposited in offshore environments clearly indicates a deepening of the depositional environments as a consequence of the early Albian transgression from the south-west through the Burgundy high (Pictet et al., 2018). This transgressive trend is also expressed by the occurrence of cosmopolitan ammonites indicating the opening of the Burgundy threshold (Fig. 1).

The third borehole, AUB240 (Fig. 2), shows a similar lithological succession but the upper part of the borehole has permitted the recovery of the topmost part of the Argiles tégulines de Courcelles and the lowermost part of the Marnes de Brienne (Fig. 7). The recognition of marker beds between the three boreholes allows precise correlations to be drawn (Amédro et al., 2019). In the upper part of this borehole, all ammonite zones were

recognized except the *Dipoloceras cristatum* Zone which is condensed and corresponds to the occurrence of a phosphate nodule bed (Amédro et al., 2019).

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# 3.2. The Col de Pré-Guittard section (Vocontian Basin)

The Col de Pré-Guittard section (Fig. 8) shows the lower to middle Albian sedimentary succession, which is part of the Blue Marls Formation. It mainly consists of hemipelagic blue-grey marls punctuated by several marker beds and bundles of micritic limestones used as marker horizons to refine the correlations throughout the basin (Friès, 1987; Bréhéret, 1997). Organic matter-rich layers (black shales) are also common, the most prominent of these being the Paquier horizon corresponding to the sedimentary expression of Oceanic Anoxic Event (OAE) 1b (Bréhéret, 1985 a and b; Tribovillard and Gorin, 1991; Hart et al., 1996; Erbacher et al., 1999; Herrle et al., 2003b). The section has been measured from the Aptian/Albian transition (Kilian bundle) to the lowermost middle Albian. Samples were taken every metre. Because different ammonite biozonations (Boreal vs Tethyan) are used in sections from the Paris Basin and the Vocontian Basin, correlations are difficult, however a comparison between the ammonite biozones used in both study areas is given in Table 1. Notably, the D. mammillatum Zone used in the Vocontian Basin includes the S. kitchini, C. floridum, H. puzosiana, Otohoplites subhilli and Hoplites (Isohoplites) steinmanni zones. During the Albian, the Paris Basin belonged to the Northern European Faunal Province of the Boreal Domain, characterized by the presence of rich populations of Hoplitidae that followed each other (including genera Sonneratia, Hemisonneratia, Otohoplites, Hoplites, Anahoplites, and Dimorphoplites), allowing the definition of many biozones. The Vocontian Basin, located on the northern margin of the Tethyan Domain, contains more occasional and less diversified faunas at the top of the lower Albian and in the middle Albian (Douvilleiceras, then Lyelliceras and Oxytropidoceras). At the top of the lower Albian, the correlation between the two basins is based on the vertical distribution of the species Douvilleiceras mammillatum which appears just above the L. regularis Zone and disappears at the top of the Lyelliceras lyelli Zone (or Subzone). In the middle Albian, Oxytropidoceras roissyanum is known at Col de Palluel at the top of this Substage (Gale et al., 2011), but here, the absence of ammonite in the lower levels does not allow to recognise the total vertical distribution of this species. The appearance of O. roissyanum is however well documented both in northern France (Amédro, 2009) and Kent in south-east England (Young et al., 2010), from the *Hoplites dentatus* Zone. Therefore, the O. roissyanum Total Range Zone used in this paper comprises successively the

Hoplites dentatus, Anahoplites intermedius, Dimorphoplites niobe and D. biplicatus zones in the Anglo-Paris Basin.

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# 4. Methods

# 4.1. Mineralogy

A total of 400 samples were analysed using X-Ray Diffraction (XRD). After moderate grinding in a mortar, powdered samples were decarbonated with a 0.2N HCl solution. The < 2 µm fraction (clay-sized particles) was extracted with a syringe after decantation of the suspension for 95 minutes following Stokes' law; this fraction was then centrifuged. Clay residue was then smeared on oriented glass slides and run in a Bruker D4 Endeavour diffractometer with CuKa radiations, a LynxEye detector and a Ni filter with a voltage of 40 kV voltage and an intensity of 25 mA (Biogéosciences laboratory, University of Burgundy, France). Goniometer scanning ranged from 2.5° to 28° for each analysis. Three runs were performed for each sample to discriminate the clay phases: 1) air-drying; 2) ethylene-glycol solvation; and 3) heating at 490°C, as recommended by Moore and Reynolds (1997) (Fig. 9). Clay minerals were identified using their main diffraction (d<sub>001</sub>) peak and by comparing the three diffractograms obtained. The following main clay minerals were identified (Fig. 9): a R0 type illite-smectite mixed-layer (17 Å based on a glycolated run condition) referred to as smectite for the following sections; a R1 type illite-smectite mixed-layer (around 11.5 Å in air-drying conditions and 13 Å after ethylene-glycol solvation); chlorite (14.2 Å, 7.1 Å, 4.7 Å and 3.54 Å peaks); illite (10 Å, 5 Å, 3.33 Å peaks); kaolinite (7.18 Å and 3.58 Å peaks) and vermiculitic clays including true vermiculite (14 Å in air-drying conditions, 14.5-15 Å after ethylene-glycol solvation and 10 Å peaks after heating) and chlorite-vermiculite mixed-layers. The relative proportions of clay minerals are estimated using peak intensity ratios, the most relevant of these being the ratio between the 7.18 Å and 17 Å peaks corresponding to kaolinite + chlorite/smectite (K+C/S). The error of this method is approximatively  $\pm$  5 %. This ratio can be taken as an estimate of the humidity vs aridity.

Fifteen samples have been measured for CaCO<sub>3</sub> content on the AUB 111 borehole with a Bernard's calcimeter device (uncertainty of ~1%). No measurement were done in the Vocontian Basin as Bréhéret, (1997), previously performed it. We only compared the GRS signal that we measured on the field with the CaCO<sub>3</sub> content given by Bréhéret (1997).

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SGR measurements were taken on the Col de Pré-Guittard section every 50 cm at the same position as for the samples collected for XRD. Field SGR measurements were taken with a constant 1-min acquisition time using a Geofyzica-Statis-Geo GS-512 spectrometer with a Cesium 137 scintillation detector. For all readings, the detector was placed against outcrop surfaces that had been cleaned of weathered material and smoothed. A scintillometer arrangement was used to produce an energy spectrum for the natural gamma rays emitted from an outcrop. This spectrum was interpreted electronically to yield concentrations of the principal gamma ray sources involved, namely potassium (K), thorium (Th) and uranium (U). K and Th are usually concentrated in clay minerals whereas U is associated with organic matter (Myers and Wignall, 1987). SGR values are given in ppm (eq. U). Measurement reproducibility is assessed at 5% (1σ) by repeating the same measurement 30 times (Martinez et al. 2013). The spectral gamma ray is an effective tool for the preliminary assessment of the degrees of palaeoweathering (clay content) and is therefore commonly used for palaeoclimate reconstructions (Ruffell et al., 2016).

#### 5. Results

#### 5.1. Paris Basin

In the Argiles tégulines de Courcelles unit, the CaCO<sub>3</sub> proportions are between 10% and 15% except in some marly limestones where the CaCO<sub>3</sub> content reaches 40%. Smear slides performed on some clayey samples show abundant nannofossils which are likely responsible for the CaCO<sub>3</sub> content. Consequently, clay minerals represent a significant amount of the deposits.

# Clay mineralogy

The clay mineral assemblages of the AUB111 borehole are mainly composed of illite (28% on average), vermiculitic clays (10% on average), smectite (22% on average) and kaolinite (26% on average). They also contain chlorite ( $\leq 5\%$ ), R1 type illite-smectite mixed-layers and chlorite-vermiculite mixed-layers, which are both estimated to be 5% or less (Fig. 3). The composition of the mineralogical assemblages along the borehole allows to distinguish three different intervals. At the base of the borehole, from 69.10 to 63.96 m, the first poorly-dated interval is characterized by abundant smectite (up to 70%), illite, and very low proportions of kaolinite. Some illite-rich samples occur in the glauconitic interval, close to the transition with the greensands. The presence of glauconite in the deposits is confirmed by the observation of smear slides. Above, the second interval is marked from 63.96 to 13.35

m by increasing proportions of kaolinite and illite together with vermiculitic clays in significant proportions (up to 15%), while the smectite proportion decreases. In this interval, the proportions of illite generally range between 20 and 30%, with occasionally more abundant proportions reaching 40% (Fig. 3). The disappearance of vermiculitic clays and the higher proportions of smectite (~ 40%) characterize the third interval, above 13.35 m while the percentages of kaolinite and illite decrease. The transition from the second to the third interval occurs around the transition from the *A. intermedius* to *D. niobe* ammonite zones.

The clay mineralogy of the AUB220 borehole is quite similar to that described in AUB111 indicating that the clay mineralogy of the Argiles Tégulines de Courcelles is, as expected, homogeneous at a kilometer scale (Figs. 4 and 5). The increase in smectites content in the third interval is better expressed, since this borehole includes an upper part of the Argiles tégulines de Courcelles (Figs. 4 and 5).

In the upper part of borehole AUB240, a fourth interval is observed from 8.75 to 27.3 m marked by a decrease in smectite while increasing proportions of illite, vermiculitic clays and kaolinite are observed in the *pricei* Zone (Fig. 7). Therefore, within the Argiles tégulines de Courcelles, the *D. niobe/D. biplicatus* time-equivalent interval is characterized by a smectite-rich sedimentation compared to the rest of the formation. This evolution in the clay assemblages confirms the low-resolution clay mineral data published by Fauconnier (1975) and Pomerol (1986).

#### 5.2. Vocontian Basin

The clay mineral assemblages of the Col de Pré-Guittard section are mainly composed of smectite (63% on average), illite (19% on average) and kaolinite (11% on average). They also contain about 5% of chlorite (Fig. 10). Traces of R1 type illite-smectite mixed-layers and chlorite-vermiculite mixed-layers may be present ( $\leq 2\%$ ) in some samples and although they were taken into account in the calculation, they are not represented in Figure 10. The clay composition is marked by a gradual smectite decrease from 70% from the base to 60% at the top of the section. This decrease corresponds to a clear increase in kaolinite with values in the order of 5% down section to values greater than 15% above. Interestingly, as in the Paris Basin, sedimentary rocks located around the Aptian/Albian boundary are depleted in kaolinite. The proportion of kaolinite increases during the *L. tardefurcata* ammonite Zone but remains generally below 10%, while in the *D. mammillatum* Zone, the proportions of kaolinite are higher than 10%.

According to Bréhéret (1997), the CaCO<sub>3</sub> content is approximately 20% at the base of the section and increases to approximately 40% at the top, which is consistent with our SGR measurements that show a reverse long-term trend with decreasing values from the base to the top of the section (Fig. 10). This expected inverse relationship between the SGR measurements and the CaCO<sub>3</sub> content was already observed in the Aptian part of the Blue Marls Formation (Ghirardi et al., 2014).

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# 6. Discussion

# 6.1. Influence of burial diagenesis

Burial diagenesis may transform clay mineral assemblages leading to a misreading of the original paleoclimatic and paleoenvironmental signal. Before examining the environmental significance of the clay minerals, it is important to ensure that they are dominantly detrital without any significant influence of burial diagenesis.

In the Paris Basin, the post-Albian geological history includes the deposition of the Upper Cretaceous chalk, the maximum thickness of which probably never exceeded 1000 m. At least 700 m of the Cenomanian to lower Campanian chalk was preserved in the depocentre of the Paris Basin (Robaszynski et al., 2005) but the total thickness was greater since the upper Campanian to Maastrichtian chalks have been dissolved (the Maastrichtian chalk is known as reworked pebbles) and eroded during the latest Cretaceous. Consequently, the Upper Cretaceous chalk is separated from the Cenozoic deposits by a major unconformity truncating Late Cretaceous folds (Guillocheau et al., 2000). In the eastern part of the Paris Basin, the chalk series was probably thinner but this was difficult to evaluate. Detailed reconstructions of the thermal evolution of the eastern part of the Paris Basin indicate that even in the Late Jurassic, the maximum burial temperature reached by the end of the Cretaceous never exceeded 50-60°C (Pellenard et al., 1999; Blaise et al., 2014; Mangenot et al., 2018). Consequently, in the overlying Albian deposits, the temperature remained low; this is confirmed by the abundance of smectite, as these minerals are known to be particularly sensitive to burial diagenesis and are transformed into R1 illite-smectite mixed-layers and illite through illitization processes at temperatures over 60-70°C (Merriman and Frey, 1999; Lanson et al., 2009; Środoń et al., 2009; Dellisanti et al., 2010). As a result, the abundance of smectite from the lowermost part of the studied boreholes indicates that the temperatures never exceeded 60-70°C and that the clay minerals are mostly primary and detrital in origin.

In the Vocontian Basin, for the same reasons, the abundance of smectite indicates a negligible influence of burial diagenesis on the clay mineralogy. Numerous studies have

shown that in the western part of the Vocontian Basin, the influence of diagenesis on clay minerals from Lower Cretaceous formations is weak (Deconinck and Chamley, 1983; Ferry et al., 1983; Deconinck and Debrabant, 1985, Levert and Ferry, 1988). In the Col de Pré-Guittard section, Rock-Eval analyses performed on organic matter-rich horizons show that  $T_{max}$  values are comprised between 417 and 437°C indicating that organic matter is immature or at the onset of the oil window, thereby confirming that the influence of thermal diagenesis remained weak on this section (Bréhéret, 1997).

# 6.2. Authigenic clay minerals

Detrital clay assemblages may be modified by the authigenesis of clay minerals formed either during early or late successive diagenetic steps. Authigenic glauconite (ironrich illite) is common in Albian deposits notably at the transition between the greensands and the Argiles tégulines de Courcelles that is at the base of the boreholes studied in the Paris Basin. Although glauconite granules are much coarser than the clay fraction, after grinding the samples, glauconitic illite is inevitably partly incorporated in the clay fraction and could be responsible for some abnormally high proportions of illite thus corresponding to a mixture of detrital and authigenic minerals (Fig. 3). This likely occurs at the base of the boreholes at the transition between the Sables verts de l'Aube and the Argiles tégulines de Courcelles.

A detailed study of clay minerals from similar Albian deposits (Gault clay) from the Wissant section in the Boulonnais area (Northwest of the Paris basin) shows that the clay fraction includes two types of smectites corresponding to beidellite (dioctahedral Al-Fe smectite). The first type, showing a fleecy shape under transmission electron microscope (TEM), is considered to be detrital, while the second displays a lathed shape due to the development of crystalline overgrowths indicating authigenesis (Holtzapffel, 1985; Steinberg et al., 1987). It is hypothesized that crystalline overgrowths are the result of an early diagenetic recrystallization of detrital smectites and may represent an early stage of glauconitization. Lath-shaped smectites are widespread in Meso-Cenozoic deposits (Holtzapffel al., 1985; Deconinck and Chamley, 1995) and result from dissolution/recrystallization processes in relatively closed environments (Clauer et al., 1990). However, the development of this type of authigenic minerals does not seem to significantly modify the composition of the detrital clay fractions.

In both studied areas, we therefore consider that the clay minerals are mainly detrital and originated from the erosion of nearby emerged lands, knowing that 1) glauconitic illite may be associated with detrital illite, notably in the Paris Basin at the transition between the

Sables Verts de l'Aube and the Argiles tégulines de Courcelles and 2) authigenic smectite may have marginally enhanced the total proportion of smectites in specific closed environments.

# 6.3 Origin of clay minerals

Although clay particles may be transported over long distances (Chamley, 1989), during the Albian, a large part of the clay minerals potentially originated from the landmasses composed of the French Massif Central and the Armorican massifs, and from the London-Brabant Massif which were the main emerged lands around the studied areas (Fig. 1). Clay minerals were likely transported to the sea mainly by rivers and then deposited by decantation relatively far from the shoreline. In the Paris basin, the erosion of the two massifs probably fed the sedimentation, whereas the clays from the Vocontian Basin had to come from the basement and the sedimentary cover of the French Massif Central and other emerged lands located to the south (Corso-Sarde microcontinent?, Fig. 1).

The lithology of the London-Brabant Massif and the French Massif Central is different. These massifs are both inherited from the Variscan orogeny, but magmatic and high-grade metamorphic rocks (granitoids, migmatites, and gneiss) are predominant in the French Massif Central, whereas low-grade metamorphic and sedimentary rocks dominated in the London-Brabant Massif (schists, slates, limestones and shales; Fielitz and Mansy, 1999; Han et al., 2000). Therefore, a different nature can be expected from the contributions of clay minerals originating from these two massifs. In addition, they were not located at the same latitudes, and according to the estimated precipitation rates during the 'Mid-Cretaceous' (Ufnar et al., 2002; 2004), the French Massif Central was located between 25 and 30°N close to an arid to semi-arid climatic belt extending southward toward Portugal and West Africa (Fenner, 2001; Heimhofer et al., 2008), while the London-Brabant Massif was located between 30 and 35°N in a more humid tropical belt.

# 6.3.1. Primary clay minerals

Primary clay minerals, i.e. chlorite and illite, are mainly formed through weathering processes of metamorphic and igneous rocks yet these minerals can be also reworked from a sedimentary basement (Chamley, 1989; Weaver 1989). In Albian deposits, chlorite never exceeds 5% of the clay fraction which is common in Cretaceous sedimentary rocks that have

not undergone a deep burial (Deconinck et al., 1985; Kühn et al., 2001). The scarcity of chlorite is explained by the overall warm and humid climate that prevailed at that time, as chlorite is easily weathered under such climate conditions (Deconinck et al., 1985). Illite primarily characterizes the mechanical erosion of a mica-rich crystalline basement (notably muscovite). The common occurrence of significant amounts of this mineral is explained by its reworking from the Variscan igneous and metamorphic massifs. The higher illite abundance in the Paris Basin (close to 30%) than in the Vocontian Basin (close to 20%) likely results from the differential settling processes of clay minerals: illite is preferentially deposited nearshore whereas smectites, because of their better buoyancy, are exported further out in the ocean (Gibbs, 1977; Chamley, 1989). This difference is then explained by the distinct palaeogeographical context of the two sedimentary basins, as the Col de Pré-Guittard Formation was likely in a more distal position compared with the emerged lands than the boreholes located in the eastern part of the Paris Basin.

# 6.3.2. Origin of kaolinite

There are two possible origins of kaolinite in the deposits. Kaolinite may originate from soils where it accumulated due to the partial hydrolysis of silicates including feldspars and other minerals which can be easily weathered (Chamley, 1989); it is subsequently transported and integrated into marine sediments. Kaolinite may be also reworked from older kaolinite-bearing sedimentary rocks or palaeoalterations (Deconinck and Vanderaveroet, 1996; Thiry, 2000). In the first case, kaolinite is indicative of a warm and humid climate responsible for strong hydrolysis, whereas in the second case, kaolinite is only indicative of a significant runoff on the continents. In both cases, the occurrence of kaolinite indicates a humid climate (Chamley, 1989; Ruffell et al., 2002; Dera et al., 2009; Bougeault et al., 2017). However, when kaolinite is reworked from ancient rocks, it can be expected to observe a covariation with illite as this mineral results from the same process, whereas when kaolinite originates from soils, its evolution through time should be decoupled from that of illite.

A clear covariation of illite and kaolinite is observed in the Paris Basin, suggesting that kaolinite is dominantly reworked from kaolinite-bearing rocks and palaeoalterations rather than from soils formed at the time of sedimentation. After the Purbeckian (latest Jurassic/earliest Cretaceous) regressive facies (Brigaud et al., 2018), the Paris Basin corresponded to a continental area where early Cretaceous Wealden facies accumulated. At that time, the surrounding massifs, remnants of the Variscan orogeny, were deeply weathered with the formation of kaolinite-rich soils, saprolites and bauxites over the French Massif

Central, the London-Brabant Massif and the Armorican Massif (Thiry et al., 2005). Apatite fission-track thermochronology performed on the Ardennes Massif suggests that the sedimentary cover of the London-Brabant Massif was eroded during the Late Jurassic and Early Cretaceous (Barbarand et al., 2018). A major alteration phase favoured by a tectonic rejuvenation in the southern part of the London-Brabant Massif took place during the Early Cretaceous and then this massif underwent a kaolinitic alteration (Quesnel, 2003; Yans, 2003; Thiry et al., 2005; 2006). In agreement with the Mid-Cretaceous study of the Isle of Wight (Gale et al.,1996; Ruffell and Garden, 1997), we suggest that clay minerals from the Albian deposits of the Anglo-Paris Basin preferentially originated from the north (i.e. the London-Brabant Massif). However, Lower Cretaceous bauxitic profiles have also been identified in the French Massif Central (Thiry et al., 2005), which could constitute an additional source of reworked kaolinite. Therefore, the high proportions of kaolinite in the Albian deposits of the Paris Basin could be explained by the erosion of the kaolinite-rich weathering profile developed over the Variscan massifs during the Early Cretaceous. As detrital clays including kaolinite may be reworked from older rocks or from palaeoalterations, their climatic significance may be questioned (Jeans et al., 2001), but overall, the illite and kaolinite dominated clay assemblages of the Albian in the Paris Basin point to a significant runoff and consequently humid climate conditions.

In the Vocontian Basin, kaolinite proportions increase slightly from the base to the top of the section, whereas the proportions of illite remain stable. This suggests that kaolinite and illite deposited in the Vocontian Basin primarily originated from distinct sources. Illite likely originated from the crystalline basement whereas kaolinite was probably at least partly derived from soils or palaeoweathering profiles developed over the French Massif Central or other further away areas. These mineralogical features suggest slightly increasing humidity during the early Albian.

# 6.3.3. Origin of smectite

Smectites seem predominantly detrital, whether in the Albian in the Boulonnais area or southern England (Holtzapffel, 1985; 1987; Gale et al., 1996). Their Al-Fe beideillite composition suggests a reworking from soils developed on the Variscan massifs under a warm and seasonally humid climate (Chamley, 1989). These minerals are abundant in a poorly-dated interval likely corresponding to the *L. tardefurcata/L. regularis* ammonite Zone in the lowermost part of the Argile tégulines de Courcelles (Figs. 3-5). Smectites are also abundant in the uppermost middle Albian corresponding to the *D. niobe* and *D. biplicatus* ammonite

zones (Figs. 3-5, and 7). During these intervals, smectite-rich sedimentation suggests that the semi-arid climatic belt expanded northward.

#### 6.3.4. Origin of vermiculitic clays

In the Paris Basin, the Argiles tégulines de Courcelles contain significant amounts of vermiculitic clays, in association with illite and kaolinite whereas these minerals are almost absent from the Vocontian Basin. These clay minerals are relatively unusual in Mesozoic marine sedimentary rocks, although vermiculite is also mentioned in coeval middle Albian deposits (Gault Clay) from the Isle of Wight in England (Gale et al., 1996). Therefore, vermiculitic minerals are likely deriving together with illite and kaolinite from a source that could possibly be the London-Brabant Massif (Fig. 11A). This mineral may originated from soils formed under cold-temperate climate (Li et al., 2019), but such climatic conditions seem unlikely at relatively low latitudes during the Albian. We therefore suggest that vermiculitic clays are reworked from the basement (moderate weathering of chlorite) or from palaeoalterations.

# 6.4. Environmental controls of the early/middle Albian clay sedimentation

The mineralogical composition of the clay assemblages is the result of a complex interplay of the lithology of the emerged landmasses, their reliefs and tectonic instability, sealevel fluctuations and climate (e.g., Chamley, 1989).

In the Paris Basin, clay sedimentation largely dominated by illite and kaolinite suggests that the London-Brabant Massif was submitted to significant runoff conditions. Between 30 and 35°N, corresponding to the palaeolatitudes of the London-Brabant Massif, precipitation rates are estimated between 1000 and 1500 mm/yr (Ufnar et al., 2002), which is high enough to produce erosional soil processes, the erosion of Lower Cretaceous kaolinite-rich palaeoalterations and the bedrock. These conditions were probably responsible for a significant detrital flux of clay minerals such as illite and kaolinite (Fig. 11). As the temperatures were also high (>20°C for these palaeolatitudes, based on  $\delta^{18}$ O values; O'Brien et al., 2017), chlorite was likely rapidly transformed into vermiculite and interstratified minerals including chlorite-vermiculite which are identified in small quantities in the Albian sedimentary rocks (Herbillon and Makumbi, 1975; Proust et al., 1986; Righi et al., 1993; Banfield and Murakami, 1998).

The two smectite-rich intervals identified in the Albian of the Paris Basin, the first one close to the Aptian/Albian transition and in the lowermost Albian and the second from the A. intermedius to the D. biplicatus ammonite zones in the lower middle Albian, clearly indicate that the erosional processes were less intense. A decreasing intensity of erosion may be linked to rising sea level or to a shift from humid to more arid conditions. At the base of the succession in the lower Albian, the long-term transgressive trend may be responsible for the smectite-rich sedimentation, but this trend lasted during the entire Albian (Haq, 2014) while abundant illite and kaolinite replaced the smectite-rich sedimentation. The comparison of clay assemblages with short-term sea level curves (Haq, 2014) did not show any relationship between clay assemblages and sea level fluctuation. In addition, in the study area, the timeequivalent interval for the D. niobe and D. biplicatus zones is known to correspond to a low relative sea level (Amédro and Matrion, 2014). Consequently, smectite-rich sedimentation does not seem to be the result of a rising sea level. However, these particular intervals, previously identified in the Boulonnais area (Holtzapffel 1985; 1987) and in southern England (Isle of Wight, Gale et al., 1996) can be explained by more arid climate conditions. As the Aptian-Albian transition is known to be a cool interval (Millán et al., 2014 and references therein), the hydrological cycle was probably slower during this period which is consistent with dryer conditions reflected by smectite-rich sedimentation intervals. Such climate conditions are also suggested in southern Portugal (Algarve) where mineralogical and palynological data indicate more aridity (Heimhofer et al., 2008). The smectite-rich sedimentation in the upper part of the middle Albian (A. intermedius to D. biplicatus zones) may be also explained by a semi-arid climate and a cooling trend suggested by calcareous nannofossils and the isotope chemistry of the Gault Clay from south-eastern England (Kanungo et al., 2018). Such climate conditions are also known in northern Spain before more humid conditions occurred during the late Albian (Sender, 2005; Coiffard et al., 2007). In the Lusitanian Basin, in the time-equivalent interval, coupled palynological and clay mineral data indicate drier conditions characterized by the abundance of Classopollis and by decreasing proportions of kaolinite balanced by increasing proportions of illite-smectite mixed-layers (Heimhofer et al., 2012).

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In the Vocontian Basin, the Aptian/Albian transition is also characterized by kaolinite-depleted deposits which is consistent with drier conditions (Fig. 11B). The proportions of this mineral slightly increased during the *L. tardefurcata* ammonite Zone but stayed relatively low. A significant increase occurs in the *D. mammillatum* Zone, partly time equivalent to the

O. subhilli and H. steinmanni zones from the Paris Basin, which represents the upper half of the D. mammillatum Zone (see Table 1 and Fig. 11B). This trend is comparable with the increase of kaolinite recorded in the Paris Basin (Fig. 11B) and likely reflects wetter climatic conditions as a consequence of the emergence of a humid equatorial belt during the Albian (Chumakov et al., 1995). There may be several possible explanations for the significant difference between the percentages of kaolinite in the Paris Basin (between 25 and 30%) and Vocontian Basin (between 10 and 15%). It is well-known that differential settling processes of clay minerals favour the nearshore deposition of kaolinite whereas smectite, because of its better buoyancy, is deposited further out in the ocean (Deconinck et al., 1985; Godet al., 2008). In addition, clay minerals deposited in the Vocontian Basin may originated from distinct detrital sources, some of which are located southward (Corso-sarde micro-continent?) in the semi-arid climatic belt where smectite should form in soils.

Surprisingly, clay assemblages do not display any particularity in the stratigraphic interval encompassing the organic matter-rich Paquier horizon (OAE1b) in the Col de Pre-Guittard section. However, the K+C/S ratio increases slightly around this level. Most works show that the temperature increased sharply during this event and some authors suggest that the monsoonal climate enhanced the humidity and detrital supply during this peculiar event (Herrle et al., 2003a and b; Hofmann et al., 2008; Jenkyns, 2010; Sabatino et al., 2015), which is consistent with our clay mineralogical results.

In the Paris Basin, the upper Albian deposits (*pricei* Zone) again show a kaolinite- and illite-rich clay fraction (Fig. 6) indicating a return to more humid climate conditions. The base of the late Albian is characterized by a warm event recorded during the *D. cristatum* Zone (Kanungo et al., 2018). Unfortunately, this stratigraphic interval is condensed in the boreholes studied here, but the above mentioned warming event (Kanungo et al., 2018) may be responsible for an accelerated hydrological cycle and consequently enhanced erosional processes on the London-Brabant Massif during the overlying *M. pricei* Zone. Alternatively, the tectonic instability of the London-Brabant Massif during the early late Albian (Owen, 1971) could be also responsible for the concomitant increase in illite and kaolinite.

**Conclusions** 

The clay mineralogy of the Albian deposits in the Paris and Vocontian basins shows some common features suggesting at least a regional climatic control of the clay sedimentation as well as specific characteristics attributable to certain local conditions.

The kaolinite-depleted interval around the Aptian/Albian transition and in the lowermost Albian suggests that a relatively cool and dry climate was likely responsible for a slow-down of the hydrological cycle. From the *D. mammillatum* Zone in the Vocontian Basin and at least from the *H. puzosiana* to the *A. intermedius* Zone in the Paris Basin, increasing proportions of kaolinite and/or illite suggest increased precipitation rates and a more constant humid climate. This change may be the consequence of the re-organization of the climatic belts, particularly the emergence of the equatorial humid zone favouring a warm humid belt at latitudes between 25 and 35°N. In the Paris Basin, kaolinite is primarily reworked from Lower Cretaceous palaeoalterations and from unmetamorphosed rocks outcropping on the London-Brabant Massif, whereas in the Vocontian Basin, this mineral may be also reworked from soils developed over the Central Massif and other continental masses located southwards. In both cases, increasing proportions of kaolinite are indicative of increased runoff triggered by a more humid climate and a southward shift of the mid-latitude warm humid belt.

During the late-middle Albian (at least in the *D. niobe* and *D. biplicatus* ammonite zones), the smectite-rich sedimentation in the Paris Basin suggests a latitudinal expansion of the semi-arid climatic belt which is consistent with palaeobotanical data. During the late Albian (*M. pricei* Zone), illite and kaolinite contents increase again indicating either a return to a more humid climate and/or the influence of a tectonic instability of the London-Brabant Massif.

The occurrence of vermiculitic clays restricted to the Anglo-Paris Basin suggests that this mineral originated from the London-Brabant Massif. Finally, the smectite-rich sedimentation recorded in the Vocontian Basin compared with the Paris Basin illustrates the significant role played by the differential settling of clay minerals from nearshore to offshore environments and by palaeogeography.

Finally, the warm climates of the Albian also seem to be characterized by fluctuations in humidity with significant impacts on the continental weathering processes.

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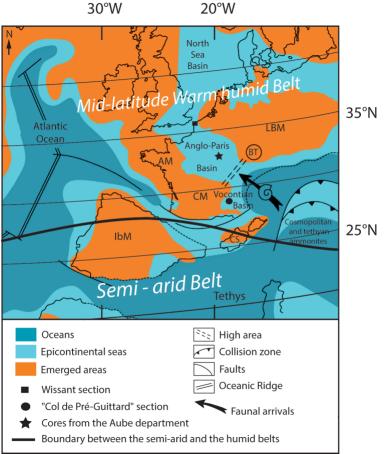
FIGURE CAPTIONS

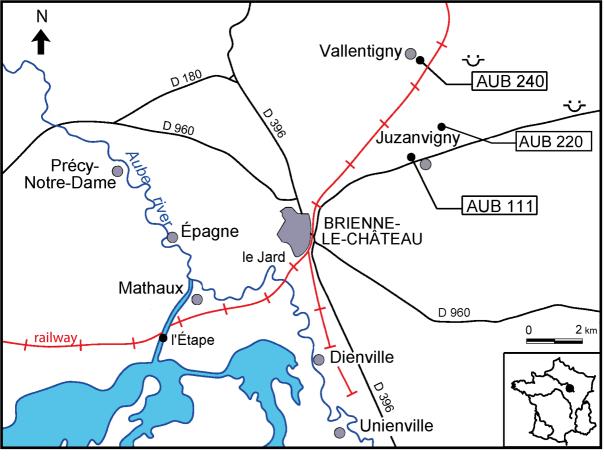
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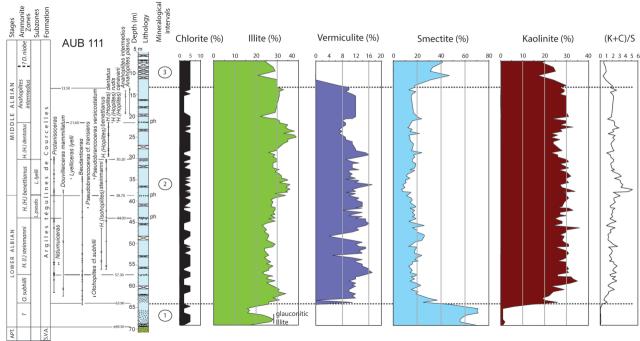
- 963 Figure 1. Palaeogeographic map (Albian) showing the location of the studied areas and the
- boundary between the semi-arid and the mid latitude warm-humid belts defined by
- Chumakov et al. (1995). Abbreviations: AM = Armorican Massif, BT = Burgundy
- threshold, MC = Massif Central, IbM = Iberian Massif, LBM = London-Brabant
- 967 Massif, CS = Corso-Sarde microcontinent (modified from Amédro and Matrion,
- 968 2014).
- 969 Figure 2. Location of the boreholes studied to the south-east of the Paris Basin (Amédro et al.,
- 970 2019).
- 971 Figure 3. Lithology, biostratigraphy and clay mineralogy of the AUB 111 borehole.
- Abbreviations: SVA = 'Sables Verts de l'Aube', APT = Aptian, (K+C)/S =
- 973 (kaolinite+chlorite)/smectite.

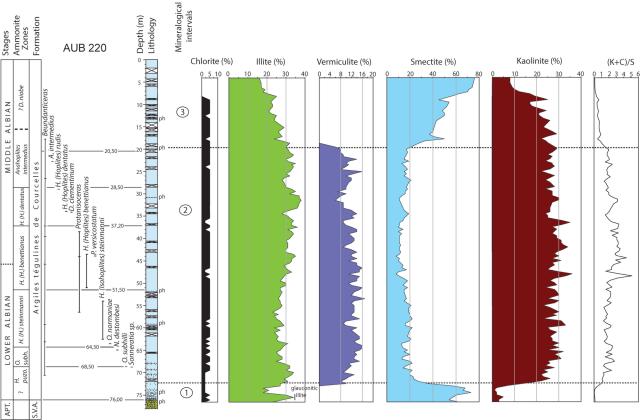
- 974 Figure 4. Lithology, biostratigraphy and clay mineralogy of the AUB220 borehole. The 975 detailed lithological description and the biostratigraphic data were published by
- 976 Amédro et al. (2017). Abbreviations : SVA = 'Sables Verts de l'Aube', APT = Aptian,
- 977 (K+C)/S = (kaolinite+chlorite)/smectite.
- 978 Figure 5. Correlation between AUB111, AUB220 and AUB240 boreholes
- 979 Figure 6. Phosphatic crust corresponding to a hardground at the transition between the 'Sables
- Verts de l'Aube' (Greensands) and the Argiles tégulines de Courcelles Formation in
- 981 borehole AUB220 (74.50 m in depth).
- 982 Figure 7. Lithology, biostratigraphy and clay mineralogy of the AUB 240 borehole. The
- 983 detailed lithological description is given in Amédro et al. (2019). (K+C)/S =
- 984 (kaolinite+chlorite)/smectite.
- 985 Figure 8. Detailed geographical location of the Col de Pré-Guittard section and field views of
- 986 this section.
- 987 Figure 9. Diffractograms obtained for the sample AUB220 25,50 m. The minerals identified
- are: S = smectite (I/S R0), V = vermiculitic clays, C =chlorite, I = illite, K = kaolinite,
- 989 C/V = interstratified chlorite vermiculite, Qtz = quartz.
- 990
- 991 Figure 10. Lithology, clay mineralogy and spectral gamma ray of the Col de Pré-Guittard
- section. (K+C)/S = (kaolinite+chlorite)/smectite.
- 993 Figure 11. A. Palaeogeographic map, facies (modified after Amédro and Matrion, 2014) and
- inferred detrital clay mineral sources in the Paris and Vocontian basins. IW = Isle of
- 995 Wight, LBM = London-Brabant Massif.
- B. Correlation between the AUB 220 borehole (Paris Basin) and the Col de Pré-
- 997 Guittard section (Vocontian Basin).
- Table 1. Correspondences between the ammonite biozonations from the Paris and
- Vocontian basins. The appearance of the foraminfer *Microhedbergella renilaevis* is
- marked in the *H. jacobi* zone. Abbreviations: U. Apt. = upper Aptian, *H. jacobi* =
- 1001 Hypacanthoplites jacobi, Micr. renil. = Microhedbergella renilaevis, L. schrammeni =
- 1002 Proleymeriella schrammeni, L. germanica = Leymeriella germanica, L. tardefurcata
- 1003 = Leymeriella tardefurcata, L. regularis = Leymeriella regularis, S. kitchini =
- Sonneratia kitchini, C. floridum = Cleoniceras floridum, H. puzosiana =
- 1005 Hemisonneratia puzosiana, O. subhilli = Otohoplites subhilli, H. steinmanni =
- 1006 Hoplites (Isohoplites) steinmanni, D. mammillatum = Douvilleiceras mammillatum,
- 1007 L.l. = Lyelliceras lyelli, L. ps. = Lyelliceras pseudolyelli, H. benettianus = Hoplites

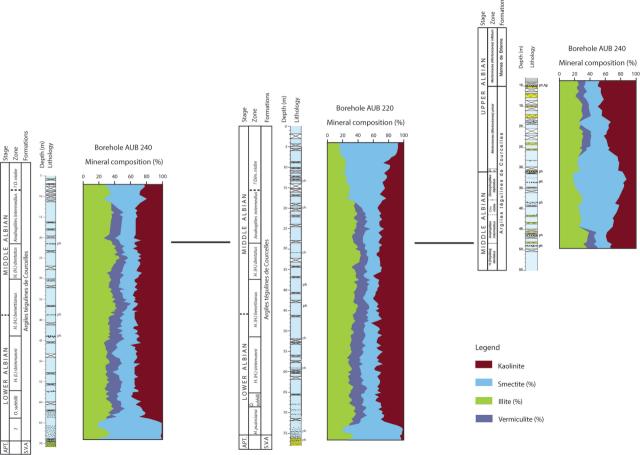
(Hoplites) benettianus, H. dentatus = Hoplites dentatus, A. intermedius = Anahoplites intermedius, D. niobe = Dimorphoplites niobe, D. biplicatus = Dimorphoplites biplicatus, O. roissyanum = Oxytropidoceras roissyanum, D. cristatum = Dipoloceras cristatum, M. pricei = Mortoniceras pricei, M. inflatum = Mortoniceras inflatum.

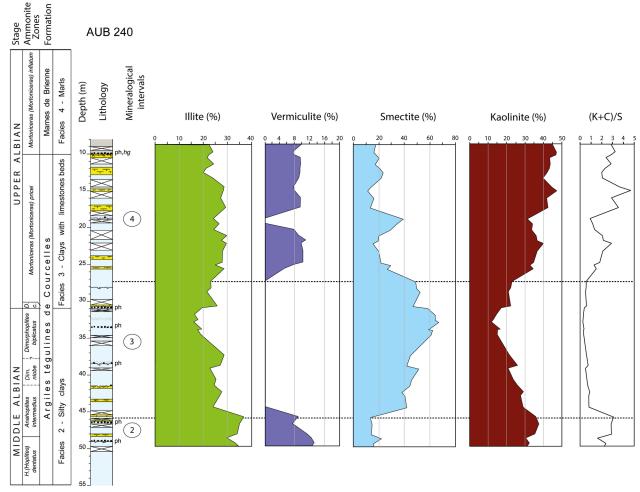


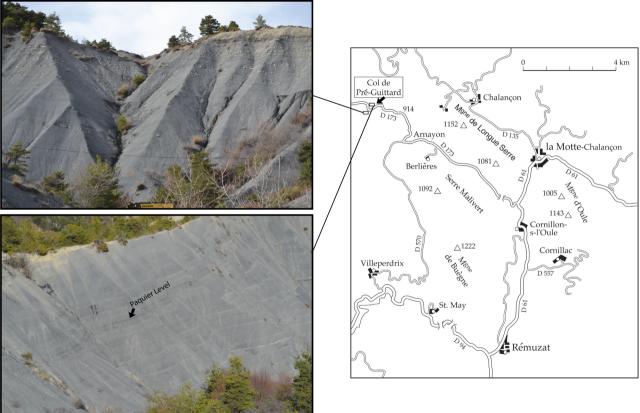


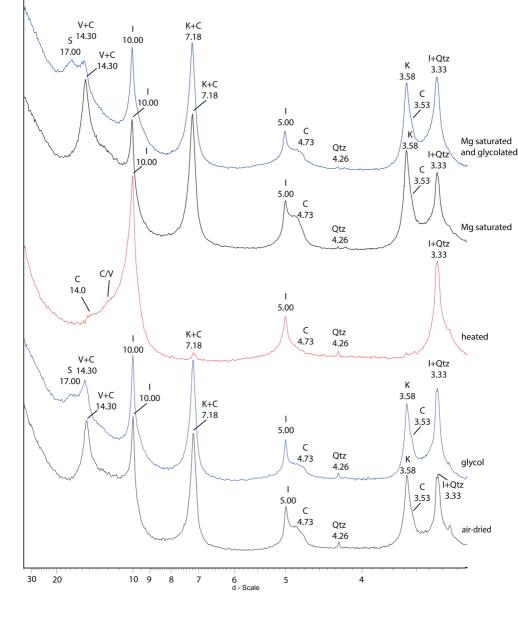


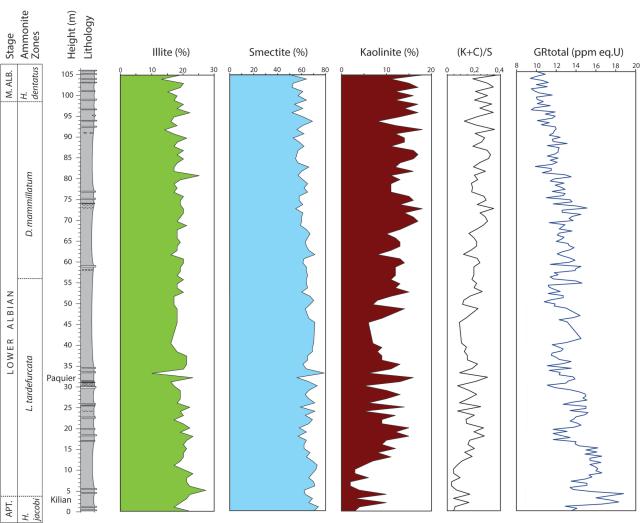


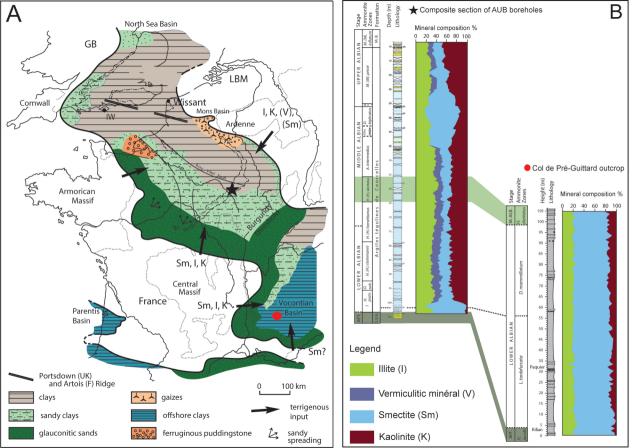












Stage	Boreal ammonite zonation (Amédro and Matrion, 2014) (Paris Basin)		Tethyan ammonite zonation (Owen, 1999; Kennedy, 2000; Gale et al., 2011) (Vocontian Basin)
Upper Albian p.p.	M. inflatum		M. inflatum
	M. pricei	ı	M. pricei
	D. cristatum		D. cristatum
	D. biplicatus		
	D. niobe		O. roissyanum
Middle Albian	A. intermedius		
	H. dentatus	Studied interval	
	H. benettianus L.l.		L. lyelli
			D. mammillatum Strdied interval
	H. steinmanni		
	O. subhilli	Stu	
	H. puzosiana	+	
Lawar Albian	C. floridum		
Lower Albian	S. kitchini		udie
	L. regularis		L. regularis ギ
	L.tardefurcata		L.tardefurcata
	L. germanica	-	L. germanica
	L. schrammeni		▲ Micr.
U. Apt.	H. jacobi		H. jacobi 🔛 renil.