

Evidence of aerial volcanic activity during the Valanginian along the northern Tethys margin

Corinne Fesneau^{a,*}, Jean-François Deconinck^a, Pierre Pellenard^a, Stéphane Reboulet^b

^a University of Burgundy, UMR CNRS 5561 Biogéosciences, 6 Bd Gabriel, 21000 DIJON, France

^b Claude Bernard University Lyon 1, UMR CNRS 5125 PEPS, Campus de la DOUA, Bâtiment Géode, 69622 Villeurbanne Cedex, France

ARTICLE INFO

Article history:

Received 4 July 2007

Accepted in revised form

18 September 2008

Available online 7 October 2008

Keywords:

Valanginian

Vocontian Basin

Volcanism

Bentonite

ABSTRACT

Stratigraphic measurement and sampling on three sections (Vergol, La Charce, and Montclus) through Valanginian deposits from the Vocontian Basin (southeastern France) reveals the occurrence of centimetre thick ochre-coloured layers, which can be correlated from one section to another. At least twelve of these are identified in sediments dated from the Pertransiens to Furcillata ammonite Zones. These horizons appear similar to previously described Oxfordian and Aptian bentonites, also from the Vocontian Basin. Clay-mineralogical and geochemical data are similar in the Valanginian ochre horizons and their enclosing marls except in one of these that shows a clay fraction mainly composed of smectite and which exhibits an enrichment in trace elements (Zr, Ba, Th, Y, Hf, U, Pb, Nb, Ta). This horizon occurs in sediments attributed to the Campylotoxus Zone and is interpreted as a bentonite while the other ochre horizons were derived from the meteoric weathering of pyrite, which probably developed in oxygen-depleted environments. Volcanic ash was presumed to be transported westward by trade winds from volcanic centres located along the northern active margin of the Tethys.

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1. Introduction

Recent investigations of pelagic deposits in three sections from the Vocontian Basin (southeastern France), Vergol, La Charce (Drôme), and Montclus (Hautes-Alpes) (Fig. 1), reveal the occurrence of twelve centimetre-thick goethite-rich horizons, which can be correlated from one section to another by their ochreous colour. These horizons were mentioned by [Beaudoin et al., \(2003\)](#) and look like previously described Oxfordian and Aptian bentonites, also found in the Vocontian Basin ([Dauphin, 2002](#); [Pellenard et al. 2003](#)). Their good correlation and facies association both suggest that they correspond to bentonites, but rigorous complementary analyses need to be achieved in order to confirm the volcanic character of these presumed bentonites. In order to characterise the ochre horizons, clay mineralogical and geochemical analyses based on immobile trace elements and rare earth elements were carried out on these layers, which were numbered O1 to O12 (Fig. 2).

During the Early Cretaceous, the Vocontian Basin was located on the northern margin of the Tethys Ocean, between 25 to 30°N ([Dercourt et al. 1993](#)). This basin was surrounded by carbonate platforms including the Jura platform to the north,

the Vivarais platform to the west and the Provençale platform to the south (Fig. 2). The La Charce, Montclus, and Vergol sections are well dated because of the abundance of ammonoids ([Atrops and Reboulet, 1995](#); [Reboulet et al. 1992](#); [Reboulet, 1996](#); [Reboulet and Atrops, 1999](#)). The biostratigraphy used here (Fig. 2) is the standard scheme adapted by the Lower Cretaceous Ammonite Working Group (Kilian Group) of the IUGS Subcommittee on Cretaceous Stratigraphy ([Hoedemaeker et al. 2003](#); [Reboulet et al. 2006](#)) for the Valanginian. All Valanginian biozones and biosubzones are considered as chronostratigraphic units further details are given by [Reboulet and Atrops, \(1999\)](#).

The sedimentary successions are dominated by heavily bioturbated marl (generally, calcareous mudrock)–limestone alternations, interpreted as resulting from nannoplankton production cycles, caused by climatic fluctuations in the Milankovitch frequency band ([Cotillon et al. 1980](#); [Giraud, 1995](#)) or related to dilution cycles by carbonate mud exported from shallow platform environments to the basin ([Reboulet et al. 2003](#)). The sediments deposited in deep-water environments are occasionally interrupted by slumped intervals of syndepositional origin and rust-coloured calcarenites showing evidence for a turbiditic origin ([Joseph et al. 1989](#); [Bulot et al. 1994](#)). These calcarenites are particularly well exposed in sediments attributed to part of the Campylotoxus and Verrucosum Zones in the La Charce section ([Reboulet, 1996](#)). The Vergol section is also characterised by the occurrence of four thin (cm-thick) layers rich in organic matter,

* Corresponding author.

E-mail address: corinne.fesneau@u-bourgogne.fr (C. Fesneau).

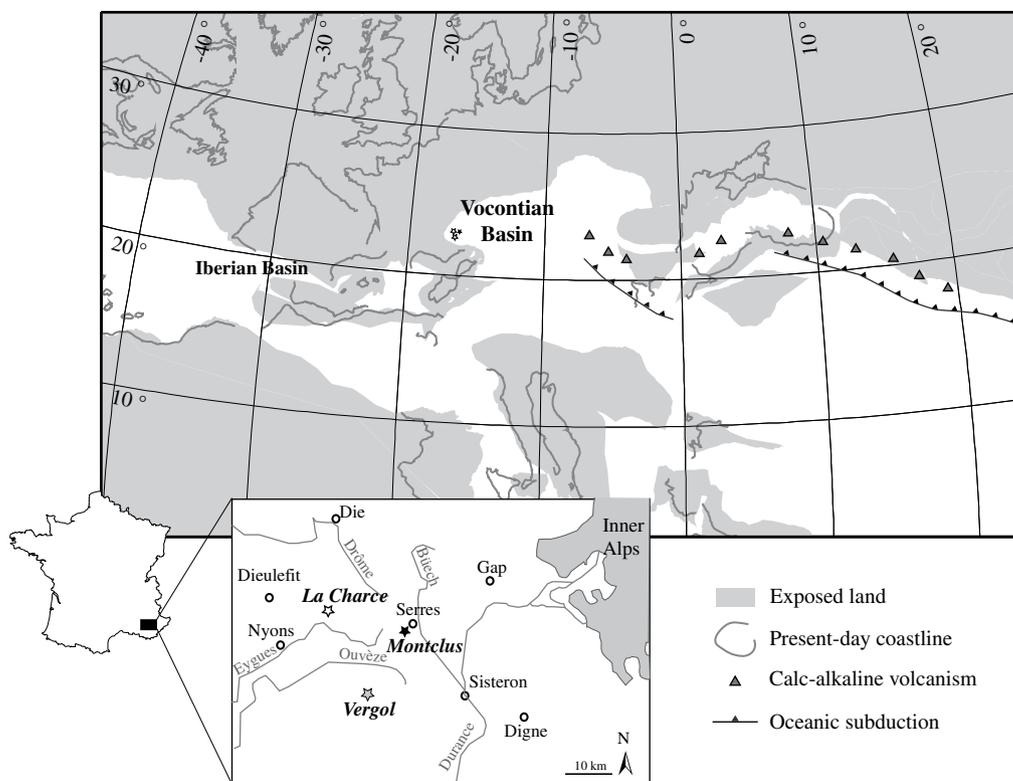


Fig. 1. Geographic and palaeogeographic location of sections studied. Map at 138 Ma modified from Geomap map generator (www.odsn.de), (Hay et al., 1999). Oceanic subduction zone and volcanism from (Dercourt et al., 1986).

called Barrande horizons, within sediments belonging to the Campylotoxus Zone, in the Biassalense Subzone (Reboulet, 2001; Reboulet, et al. 2003). Finally, from sediments dated as the top of the Pertransiens to the Furcillata Zones, at least twelve ochre horizons are identified.

2. Methods

Clay mineral associations were studied using X-ray diffraction (XRD) on oriented mounts. Each crushed sample was decarbonated using 0.2 N hydrochloric acid. Excess acid was removed by successive washing with deionised water. The clay fraction (<2 μm) was separated by settling and centrifuge. X-ray diffractograms were obtained using a Siemens D500 diffractometer with $\text{CuK}\alpha$ radiation and Ni filter. For each sample, three X-ray analyses were performed: after air-drying, ethylene-glycol solvation, and heating at 490 °C for two hours. The identification of clay minerals was made according to the position of the (001) series of basal reflections on the three X-ray diagrams (Brown and Brindley, 1980; Moore and Reynolds, 1989). Semi-quantitative analyses were obtained from the area of the (001) series of mineral basal reflections on the ethylene-glycol solvation trace using MacDiff 4.2.5 software by Petchick (2001). Clay mineral assemblages were studied in the ochre horizons and, for comparison, in the enclosing shales located a few centimetres above and below.

Geochemical data were obtained using the inductively coupled plasma (ICP) technique after LiBO_2 and HNO_3 digestion. Major elements were analysed by ICP-AES, minor elements and REE by ICP-MS (Nancy CRPG Laboratory). Ten major and 43 trace elements were determined in whole rock samples from ochre horizons and associated shales.

3. Results: clay mineralogy and geochemical data

Clay assemblages of the ochre horizons include illite, kaolinite and I/S mixed-layers. In most cases, no significant differences are observed between the clay proportions of ochre horizons and the enclosing marls, except for the O3 ochre horizon (Campylotoxus Subzone), where the clay fraction is composed of more than 90% of well-crystallised smectites with traces of kaolinite and illite (Fig. 3). The enclosing marls show a clay fraction composed of the usual terrigenous suite comprising illite/smectite mixed-layers (35%), illite (25%), kaolinite (30%) and chlorite (10%) (Deconinck et al. 1985; Levert and Ferry, 1988, Fig. 3). Therefore a significant difference in clay mineralogy exists between the enclosing marls and ochre horizon O3.

Geochemical analyses performed on ochre horizon O3 and enclosing marls reveal noteworthy differences (Table 1; Fig. 4). The profile of trace elements shows a abundance of some elements Zr, Ba, Th, Y, Hf, U, Pb, Nb, Ta and As, Mo, Sn. The profile of REE normalised to the Cody-Shale (Sco-1) shows a depletion of LREE (La, Ce, Pr, Nd and Sm) REE profiles from the other ochre horizons reveal no significant differences with their enclosing marls. The Al_2O_3 content of O3 is twice as high as that of its enclosing marls (Table 1).

4. Discussion

4.1. Origin of ochre layers

Bentonite horizons previously described in marine successions are generally characterised by abundant well-crystallised smectites derived from the submarine alteration of volcanic glass shards (Deconinck and Chamley, 1995; Meunier et al. 1999; Pellenard et al.

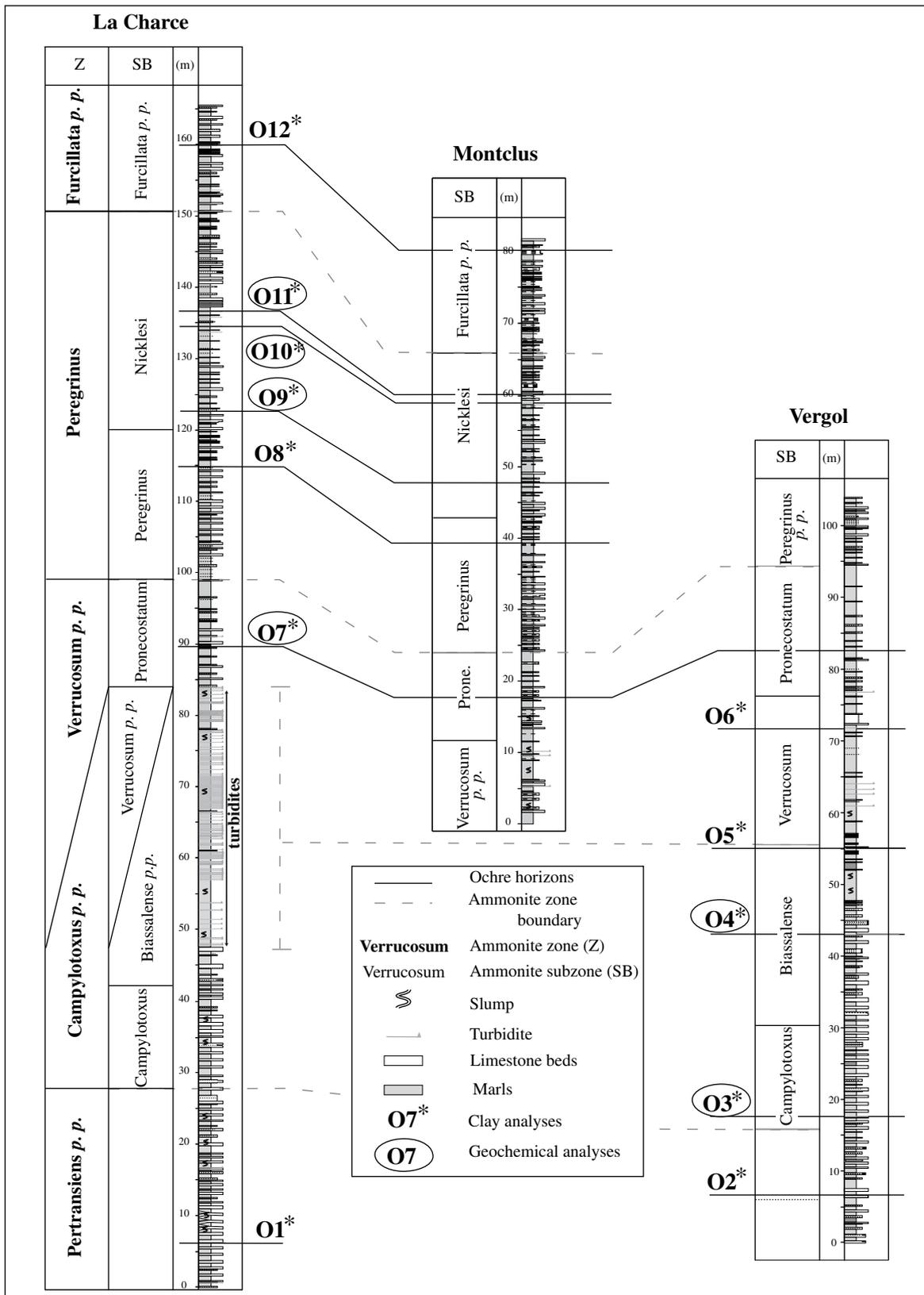


Fig. 2. Biostratigraphy of the three sections (based on Reboulet, 1996; this work) and their correlation, lithostratigraphy and ochre horizons.

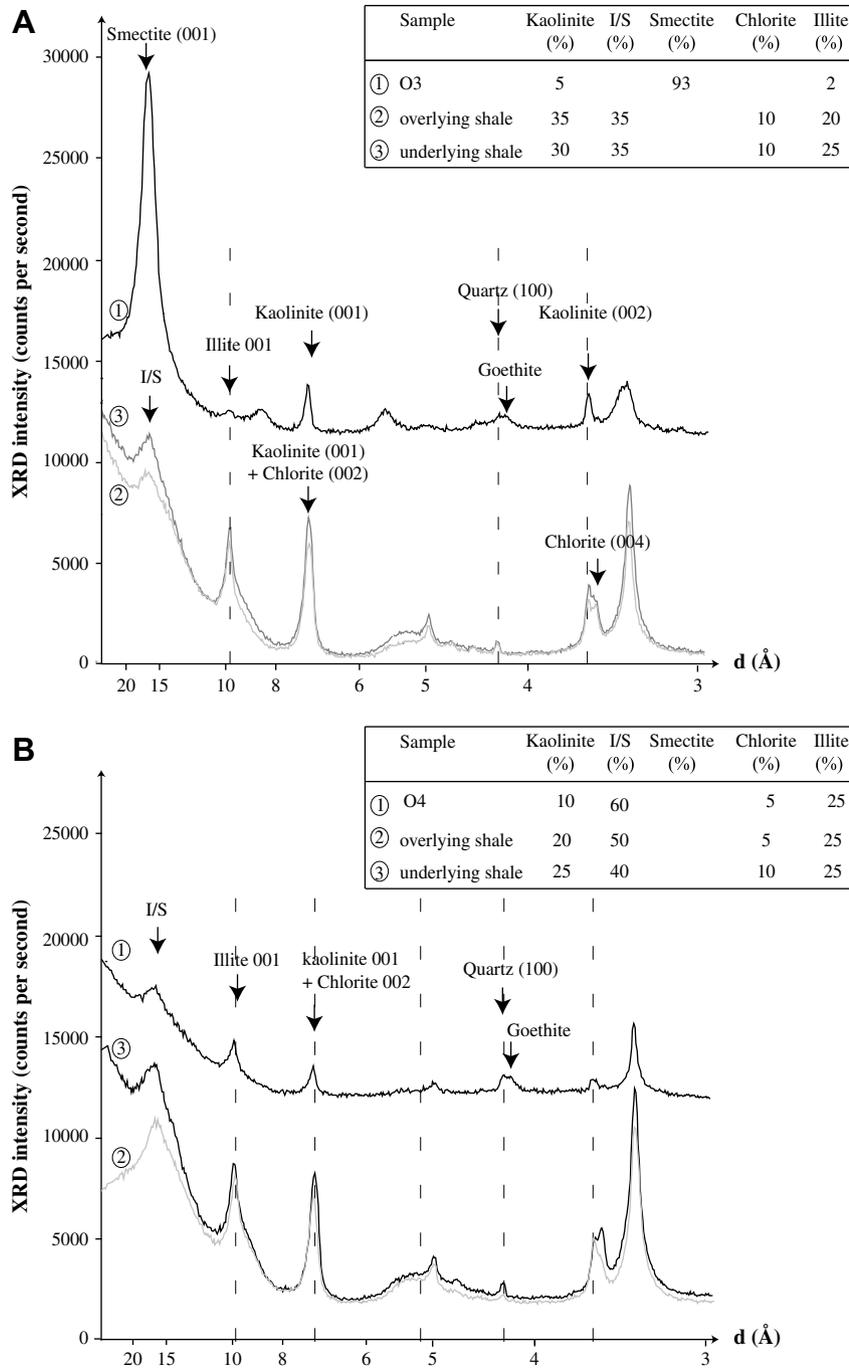


Fig. 3. A: Comparison between X-ray diffraction traces (glycol-solvated) of the clay fraction of the O3 bentonite and the clay fraction of its enclosing marls (O3 ± 5 cm). B: Comparison between X-ray diffraction traces (glycol-solvated) of the clay fraction of the O4 ochre layer and the clay fraction of its enclosing marls (O4 ± 5 cm).

2003; Wray, 1995; Wray, 1999). The abundance of these minerals in ochre horizon O3 compared to its enclosing shales (Fig. 3), and the abundance of elements with a specific magmatic affinity (Zr, Ba, Th, Y, Hf, U, Pb, Nb and Ta) (Fig. 4B), are consistent with a volcanic origin (Clayton et al. 1996; Pellenard et al. 2003; Spears et al. 1999), and confirm that O3 corresponds to a bentonite.

As there are no significant differences in clay mineral assemblages and chemical composition in comparison between enclosing marls and the remainder of the ochreous horizons, the volcanic signature of the other ochre horizons is not confirmed. These horizons cannot be interpreted as bentonites despite both their field aspect (moisture, plasticity) and the observation that

these layers are correlatable in the studied sections. These layers may have derived from the meteoric weathering of pyrite-rich horizons, probably developed in oxygen-depleted environments at the expense of organic matter through sulphate reduction processes.

4.2. Chemical fingerprint and magmatic affinity of O3 bentonite

The replacement of siliceous glass shards by clay minerals during submarine weathering at the seawater/sediment interface is responsible for chemical exchanges, particularly of mobile elements. The O3 bentonite shows a SiO₂/Al₂O₃ ratio lower than

Table 1
Major, trace element and REE analyses of the samples studied

	Overlying shale	O3	underlying shale
Major elements (wt %)			
SiO ₂	22.65	35.46	24.30
Al ₂ O ₃	7.84	15.66	8.90
Fe ₂ O ₃	2.84	28.63	2.74
MnO	0.02	0.00	0.02
MgO	1.46	1.34	1.26
CaO	33.32	1.75	32.06
Na ₂ O	0.09	0.21	0.09
K ₂ O	1.18	1.23	1.25
TiO ₂	0.35	0.80	0.38
P ₂ O ₅	0.06	0.10	0.05
SiO ₂ /Al ₂ O ₃	2.89	2.26	2.73
Trace elements (p.p.m)			
As	4.46	34.54	3.72
Ba	94.26	3054.00	99.48
Be	0.97	0.75	1.21
Bi	0.15	0.42	0.15
Cd	BD	0.34	0.38
Co	7.69	9.67	11.32
Cr	47.12	33.03	51.36
Cs	5.01	4.13	5.62
Cu	14.65	15.27	12.83
Ga	11.25	16.10	12.46
Ge	1.27	2.06	1.50
Hf	4.08	19.48	4.51
In	BD	0.17	BD
Mo	0.53	32.18	1.29
Nb	17.84	47.92	17.69
Ni	34.74	56.74	44.09
Pb	10.16	115.28	20.37
Rb	60.91	48.44	66.67
Sn	2.01	10.90	2.65
Sr	567.30	184.70	538.20
Ta	1.48	16.16	1.48
Th	5.50	38.35	6.29
U	3.10	4.22	3.79
V	60.70	57.49	68.01
W	0.72	0.91	0.85
Y	17.95	20.73	17.84
Zn	45.74	69.49	138.80
Zr	145.40	406.50	147.20
Rare earth elements (p.p.m)			
Ce	0.77	0.33	0.79
Dy	0.71	0.77	0.73
Er	0.69	0.76	0.69
Eu	0.46	0.41	0.49
Gd	0.64	0.48	0.64
Ho	0.61	0.67	0.61
La	0.80	0.35	0.82
Lu	0.82	0.79	0.84
Nd	0.71	0.31	0.73

BD: below detection.

the enclosing marls, indicating release of silica and formation of clay minerals (Pellenard et al. 2003). The Al₂O₃ content of 15.7% is the average composition of typical bentonites (Spears et al. 1999). The depletion of LREE recorded in the O3 bentonite (Fig. 4) may result from the argillation of volcanic glasses (Clayton et al. 1996; Morton and Knox, 1990; Wray, 1995).

Direct geochemical connection between bentonite and magmatic source rock is often difficult to establish because atmospheric conditions prevailing during eruption processes may induce the sorting and aeolian fractionation of ash. The geochemical composition of bentonites also depends on meteoric alteration. Consequently, geochemical data from bentonites should be interpreted with caution as weathering processes and contamination from their enclosing shales may have significantly modified the original composition of the volcanic ash-falls.

The more immobile chemical elements including Zr, Ti, Y and Nb, are generally used to determine the initial composition of the volcanic ashes and the type of magma (Pellenard et al. 2003; Spears et al. 1999; Winchester and Floyd, 1977). The discrimination diagram of Winchester and Floyd (1977) is based on Zr/TiO₂ for the alkalinity index and Nb/Y for the differentiation index, but Savostin et al. (1986) and Spears et al. (1999) have shown that this diagram must be used with caution, notably because ratios of Zr/TiO₂ and Nb/Y are highly dependent on the abundance of heavy minerals (zircon, anatase, rutile, ilmenite, and monazite). In spite of thorough examination, no volcanic minerals (such as zircon, apatite, sanidine and biotite) were found in this horizon, probably because it corresponds to a distal deposit of volcanic ash. Despite this, the diagram of Winchester and Floyd plots O3 in the trachyandesitic composition field, which is confirmed by the TiO₂-Zr crossplot from Leat et al. (1986), suggesting that the composition of the original ash derived from an intraplate volcanic setting and an alkaline series. To provide another confirmation of the identified tectonomagmatic fields, bentonite trace elements were plotted on the diagram of Wood (1980) (Fig. 5). In the Th-Hf/3-Ta triangular diagram, the O3 data are close to the field of an alkaline series characteristic of intraplate or rifting (continental or oceanic) magmatism. This was confirmed by a Tbx3-Tax2-Ta triangular diagram (Cabanis and Thiéblemont, 1988). To conclude, immobile trace element chemistry suggests a trachyandesitic composition for the original ash derived from an alkaline series in an intraplate volcanotectonic setting. The O3 bentonite corresponds to a distal ash fall and no pyroclastic minerals have been found, so the possibility of differentiation during aeolian transport and water settling cannot be ruled out.

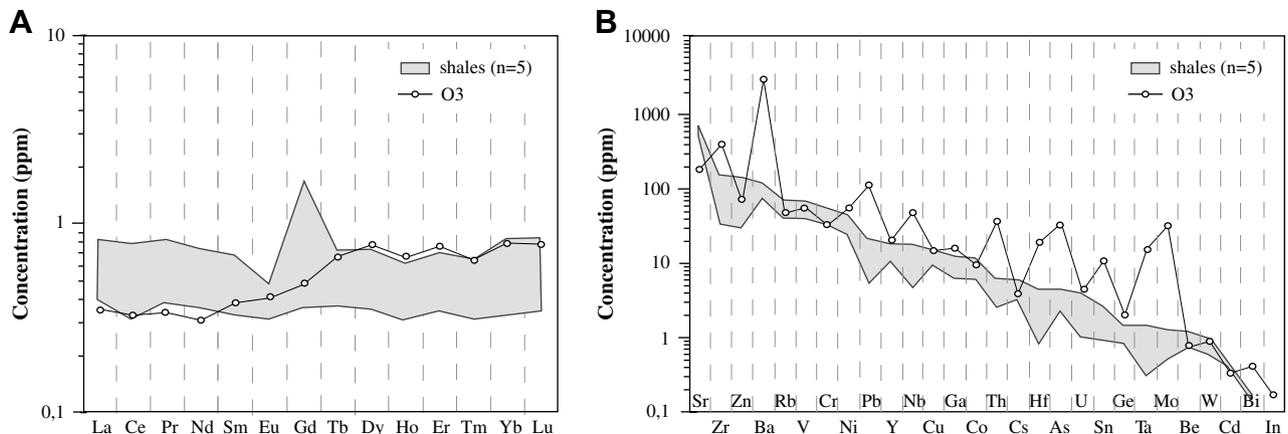


Fig. 4. Geochemistry of the O3 bentonite and associated shales from the Vocontian Basin. (A) Cody shales-normalised Rare Earth Element profiles. (B) Non-normalised trace element profiles. Cody shale values from Jarvis and Jarvis (1985).

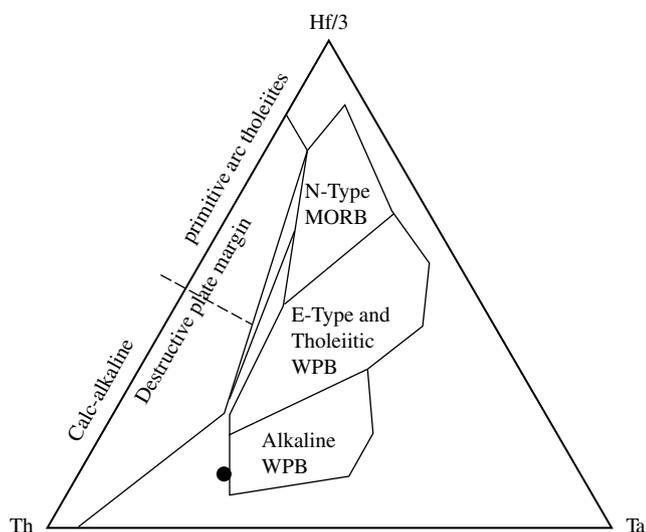


Fig. 5. Location of O3 bentonite plotted on the diagram of Wood (1980); WPB: within-plate basalts and differentiates.

4.3. Location of the active volcanic centre

The presence of an altered ash fall deposit in the Valanginian succession of the Vocontian Basin indicates that aerial explosive volcanism occurred during this period and the absence of coarse pyroclast grains, probably reflects a distal deposit from tropospheric/stratospheric clouds of volcanic particles following a Plinian or ultraplinian paroxysmal eruption. Valanginian bentonites have, to our knowledge, never been mentioned in other European basins. In the Subbetic Domain (Betic Cordillera, southern Spain) to the west (Fig. 1), sections consisting of hemipelagic deposits similar to those of the Vocontian Basin were studied in detail but no bentonites were found (Company, 1987; Hoedemaeker and Leereveld, 1995; personal field work). It is therefore currently impossible to compare bentonite thickness over long distances in the hope of finding a polarity which would indicate the location of active volcanic centres. The Early Cretaceous is characterised by intraplate volcanism associated with the rifting and opening of the south and central Atlantic (Savostin et al. 1986). This hypothesis is consistent with the geochemical data but, according to atmospheric circulation models (Poulsen et al. 1998; Price et al. 1995), volcanic ash would have been advected by eastern trade winds from a volcanic source located on the northern margin of the Tethys.

An alternative origin may be provided with volcanic activity associated with the subduction zones located to the east, on the northern margin of the Tethys Ocean. This calco-alkaline volcanism can be followed from the Roumanian Marmaros to the Sinaia unit (Dercourt et al. 1986). Its calco-alkaline affinity is, however, inconsistent with the chemical analyses of the O3 bentonite. In spite of this, the eastern origin should not be rejected as the chemical composition of the bentonite may have undergone weathering processes and contamination from its enclosing marls. Such orogenic volcanism is well known to produce intense paroxysmal eruption often associated with ultraplinian or plinian activity.

Based on all the above reasoning, the active volcanic centres associated with subduction zones located on the northern active margin of the Tethys Ocean are the most privileged origin for the Valanginian bentonite.

5. Conclusions

At least twelve thin ochre horizons that may correspond to bentonites are identified in the Valanginian deposits of the Vocontian

Basin. Some of these are correlated in three sections studied here. Mineralogical and geochemical analyses reveal that only one of these (occurring in sediments attributed to the Campylotoxus Zone) corresponds to a bentonite layer. In the field, this horizon is identifiable by its rust colour. It shows a clay fraction composed almost entirely of smectite derived from the submarine weathering of volcanic glass shards. The abundance of specific elements of magmatic affinity is consistent with a volcanic origin. According to the palaeogeographic and geodynamic settings, the volcanic ash-falls probably originated from the northern Tethyan active margin. The other ochre horizons were derived from the meteoric weathering of pyrite into goethite and should not be confused with a volcanic horizon. Their clay fraction is similar to those of the enclosing marls and their chemical characteristics are close to common shales.

Acknowledgments

The authors acknowledge Professors Karl Föllmi and Alastair Ruffell for constructive reviews and Carmela Chateau-Smith for her assistance with the English version.

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