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LA-ICP-MS dating of detrital zircon grains from the Cretaceous allochthonous bauxites of Languedoc (south of France): Provenance and geodynamic consequences.

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Abstract

The Cretaceous of southern France is characterized by a long erosional hiatus, outlined with bauxite deposits, which represent the only remaining sedimentary record of a key period for geodynamic reconstructions. Detrital zircons from allochthonous karst bauxites of Languedoc (Southern France) have been dated using LA-ICP-MS (Laser Ablation Inductively Coupled Plasma Mass Spectrometry), in order to specify the age of deposition and to constrain the provenance of the weathered material. We analysed 671 single detrital zircon grains from 3 karst bauxitic basins, stretching from close to the Variscan Montagne Noire to the present-day Mediterranean Sea. Analytical results provide Variscan (300-350 Ma) and Late Proterozoic (550-700 Ma) ages as primary groups. In addition, Middle-, Late Proterozoic and Early Archean (oldest grain at 3.55 Ga) represent significant groups. Mid-Cretaceous zircons (118-113 Ma) provide a pooled age of 115.5 ± 3.8 Ma, which constitutes the maximum age for bauxite deposition. Results also suggest a dual source for the Languedoc bauxite: one generalized sedimentary source of regional extent and a localised source in the Variscan basement structural high, that has been progressively unroofed during Albien. Integration of these new findings with previously published thermochronological data support the presence of an Early Cretaceous marly cover on the Variscan basement, which has been weathered and then removed during the Albien. The Languedoc bauxite provide a spatial and temporal link between the uplift of southern French Massif Central to the north, and the Pyrenean rift and its eastward extension to the south. These new results allow to constrain the timing and distribution of uplift/subsidence during the mid-Cretaceous events in relation with the motion of the Iberian plate relative to Eurasia.

Keywords

Karst ; Erosion ; Provenance analysis ; Durancian uplift ; Pyrenees ; Mid-Cretaceous

1- Introduction

Bauxite derives from continental weathering of aluminosilicate-rich rocks, such as crystalline basement, claystones or marls, under tropical hot and humid conditions (Tardy, Y. & Roquin, C., 1998). Some bauxites are formed as a weathering cap, especially on the surface of stable cratons, where they are known as lateritic bauxite (Bardossy, G. & Aleva, G. J. J., 1990). In Mediterranean Europe, most bauxite deposits correspond to karst bauxite (Bardossy, G., 1982 ; Herrington, R. *et al.*, 2016). These bauxites are allochthonous, and have been transported from the place where the parent rock was weathered into original lateritic bauxite (Bardossy, G. & Aleva, G. J. J., 1990; Bardossy, G. & Dercourt, J., 1990; Bardossy, G. & Combes, P.-J., 1999; Mongelli, G. *et al.*, 2015).

The reworked bauxite was trapped on the irregular surface of karstified limestones and sealed by a younger sedimentary cover. The unconformity of karst bauxite over their carbonate bedrock can be associated with a significant hiatus, corresponding to the time interval required to allow the carbonate massif to be eroded and karstified, prior to bauxite deposition. Both the erosional unconformity over the karstified bedrock and the dismantling / transport of the initial bauxite cap require some tectonic activity affecting the area, in order to ensure i) basement uplift and drainage of the weathered zone, and ii) denudation and karstification of the carbonate substratum prior to bauxite reworking and trapping. However, tectonics must remain moderate to maintain low topographic gradients that ensure optimal drainage for weathering (Wyns, R. *et al.*, 2003). The stratigraphic record of allochthonous bauxites may thus provide invaluable information about poorly documented periods characterized by stratigraphic hiatuses.

Southeastern France's geological record displays such a large-scale stratigraphic gap extending across part of, or most of, the Cretaceous (Chantraine, J. *et al.*, 1996), classically known as the « Durancian Isthmus » (Gignoux, M., 1926). This episode was characterized by erosion and karstification of platform carbonates related to the northern Tethyan margin, as well as intense weathering leading to the formation of bauxite deposits (Combes, P.-J., 1990)

Such a hiatus obliterates the understanding of a key period of the geodynamic evolution of southern Europe, when important events occurred such as : 1) rifting between Iberia and Europe, involving hyperextension, and mantle exhumation (Lagabrielle, Y. & Bodinier, J.-L., 2008) (Lagabrielle, Y. *et al.*, 2010) (Tugend, J. *et al.*, 2014); (Clerc, C. *et al.*, 2015; Cochelin, B. *et al.*, 2018; Oldum, M. L. & Stockli, D. F., 2019) (Lagabrielle, Y. *et al.*, 2019); 2) basin subsidence and deposition of thick sequences of deep marine sediments from the present-day eastern Pyrenees (Souquet, P. *et al.*, 1985) (Debroas, E.-J., 1990) (Chelalou, R. *et al.*, 2016) to offshore Provence (Fournier, F. *et al.*, 2016) and 3) the uplift of the Variscan basement of southern France (Barbarand, J. *et al.*, 2001; Séranne, M. *et al.*, 2002; Peyaud, J. B. *et al.*, 2005); (Olivetti, V. *et al.*, 2020). Correlating these different events first requires a precise chronology. Unfortunately, until now, the bauxite-related hiatus and poor age constraints have prevented such correlations.

U-Pb dating of detrital zircon grains from sedimentary formations pursue several objectives (Gehrels, G. E., 2014). The youngest detrital zircons provide the maximum deposition age for deposition of sediments (Margalef, A. *et al.*, 2016). Detrital zircons also reflect the provenance of the sediments, i.e. the uplifted areas exposed to erosion and providing terrigenous material to sedimentary basin, as for the flysch basins (Martínez, F. J. *et al.*, 2016) and late orogenic basins (Pfeifer, L. S. *et al.*, 2018) of the Variscan orogeny of southern Europe, or for the Oligocene continental rift basins in Provence (Villeneuve, M. *et al.*, 2018). When detrital zircons are found in allochthonous karst bauxite, they may effectively address the nature of the protoliths of the alterites (Boni, M. *et al.*, 2012); (Mongelli, G. *et al.*, 2016).

The aim of this study is to analyse detrital zircon grains from the allochthonous bauxites of Languedoc, by U-Pb LA-ICP-MS, in order to address the bauxites source (basement rocks vs sedimentary cover) and their provenance. In turn, this will make it possible to determine the timing and location of uplift and subsidence. The results are then discussed in the perspective of the mid-Cretaceous geodynamics in southern France. In particular, we investigate the correlation between the bauxite event in Languedoc with i) the Durancian Isthmus, ii) uplift and denudation of the southern French Massif Central, and iii) subsidence and flysch deposition along the future Pyrenees and offshore Provence.

2- Geological setting

The Variscan basement in the southern French Massif Central crops out in the Cévennes and Montagne Noire highs (Fig. 1). It consists of Precambrian to Paleozoic metasediments and para- and orthogneisses which were intensely deformed during the Variscan orogeny (Faure, M. *et al.*,

2009). In the late stage of the orogeny, they were intruded by Carboniferous post-collisional granitoids (Brichau, S. *et al.*, 2008 ; Roger, F. *et al.*, 2015) and their volcanic equivalents are preserved in coal-bearing, intra-montane basins (Bruguier, O. *et al.*, 1998; Bruguier, O. *et al.*, 2003). Following Stephanian and Permian late-orogenic extension (Echtler, H. & Malavieille, J., 1990 ; Van Den Driessche, J. & Brun, J. P., 1992 ; Faure, M., 1995), Triassic erosion formed an extensive planation surface (Bourquin, S. *et al.*, 2011). During Lower Jurassic (Lias), the area underwent NW-SE rifting, which later, led to the opening the Tethys ocean (Dubois, P. & Delfaud, J., 1989). The southern Massif Central Variscan basement of was overlapped by the very thin Trias to Jurassic transgressive sequences preserved as remnants of Triassic sandstones and Liassic littoral facies (Bonijoly, D. *et al.*, 1996), onto the post-orogenic erosional peneplain, which now appears as an exhumed paleosurface (Aubague, M. *et al.*, 1977). This Trias-Jurassic sequence is thickening – and becomes more complete – southeastwards, across the Cévennes structural high, to the “Sud-Est” Basin (Beaudrimont, A. F. & Dubois, P., 1977). Thermal subsidence of the Tethys passive margin led to deposition of thick sequences of Jurassic marine carbonates and early Neocomian marls (Fig. 2) (Beaudrimont, A. F. & Dubois, P., 1977; Mascle, A. *et al.*, 1996). The Early Cretaceous (Valanginian to Hauterivian) marls and marly limestones sequence, extended over the basement, as suggested by the thickening and deepening trend from the “Sud-Est” basin (Fig. 1) towards the present Variscan basement highs (Arnaud, H. *et al.*, 1984; Cotillon, P., 1984; Amouroux, G., 2003). The post-rift sequence is truncated by a regional erosional surface that removed part of the Jurassic to Neocomian sequence along an E-W bulge that links the Maure-Esterel Variscan high, through Provence, to the southern French Massif Central (Fig. 1), known as the “Durancian Isthmus” (Gignoux, M., 1926) or “Durancian uplift”. The erosional hiatus suggests a kilometre-scale uplift and denudation (Masse, J.-P. & Philip, J., 1976).

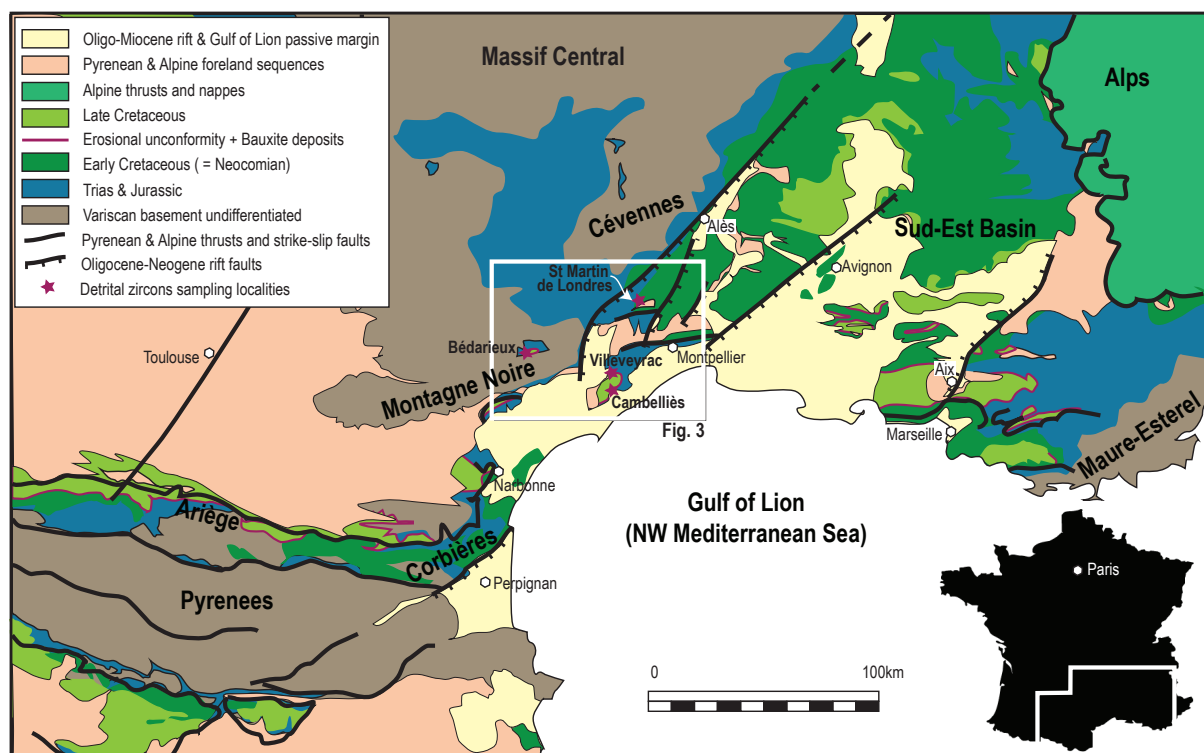


Fig. 1

Fig. 1: Structural setting of southern France, synthesised from BRGM 1/10⁶ scale map of France. The study area (corresponding to the location of Fig. 3) displays 3 bauxite basins (Bédarieux, Villeveyrac and Cambelliès) and the 4 sampling localities (plain red stars).

In the study area, north of Montpellier, Valanginian is preserved in down-faulted blocks, separated by NE-trending and EW faults (Fig. 3). The blocks display residual bauxite deposits on

their footwall or are sealed by late Cretaceous deposits (Philip, H. *et al.*, 1978) (Andrieux, J. *et al.*, 1971). Some of these east-trending normal faults have been reactivated as thrusts during the later Pyrenean orogeny (Fig. 3). Southeast of the Villeveyrac basin, bauxite deposits seal one of such faults (Gottis, M. *et al.*, 1967), clearly indicating that block-faulting predates bauxite deposition. The stratigraphic and structural relationships of such faults relate them to the Durancian uplift, which have often been reactivated as thrust and left lateral ramp during the later Pyrenean orogeny (Husson, E., 2013) (Fig. 3). The Durancian uplift is coeval with uplift of the Variscan basement in the southern French Massif Central, where apatite fission track (AFT) analyses imply an erosional denudation of up to 2000m (Barbarand, J. *et al.*, 2001; Peyaud, J. B. *et al.*, 2005).

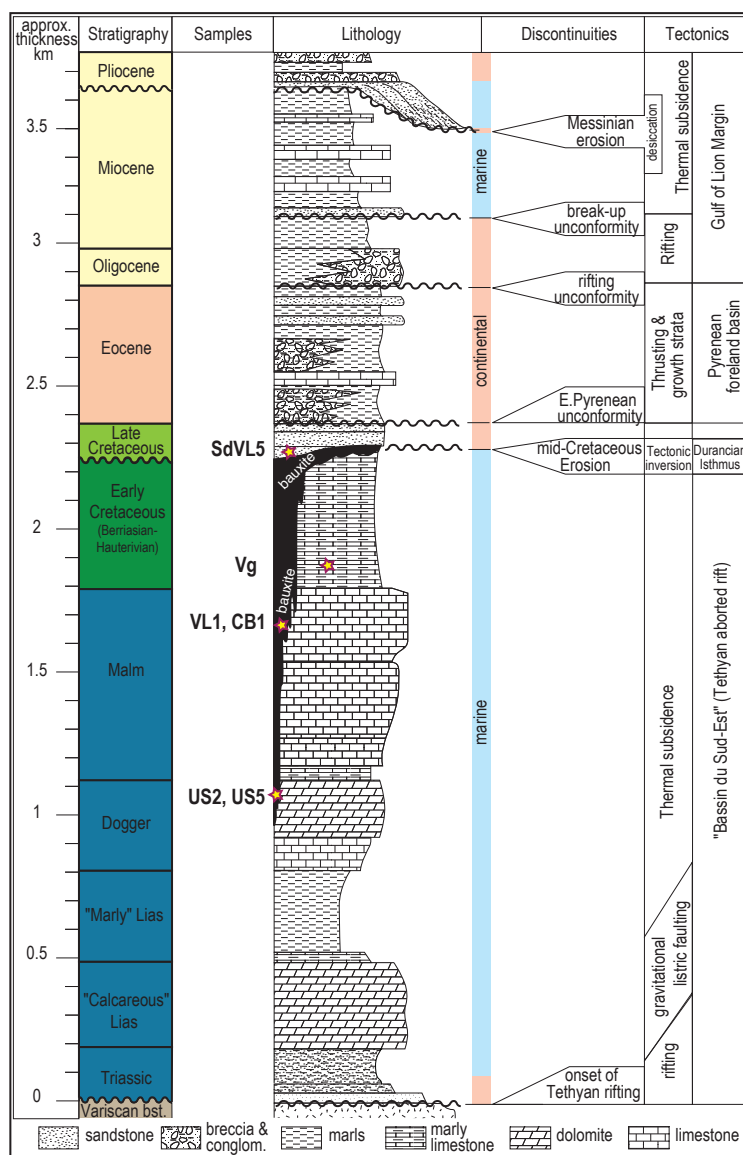


Fig. 2 : Simplified lithostratigraphic column of Languedoc correlated to the main discontinuities and tectonic events. Same stratigraphic legend as in Fig. 1. Note that bauxite deposits (in black) are highly unconformable onto the Jurassic sequence, and correspond to a stratigraphic hiatus of increasing duration from east to west. Consequently, the karst bauxite samples (US2, US5, VL1, CB1) are located relatively to the stratigraphic position of the wall (modified from (Husson, E. *et al.*, 2018)).

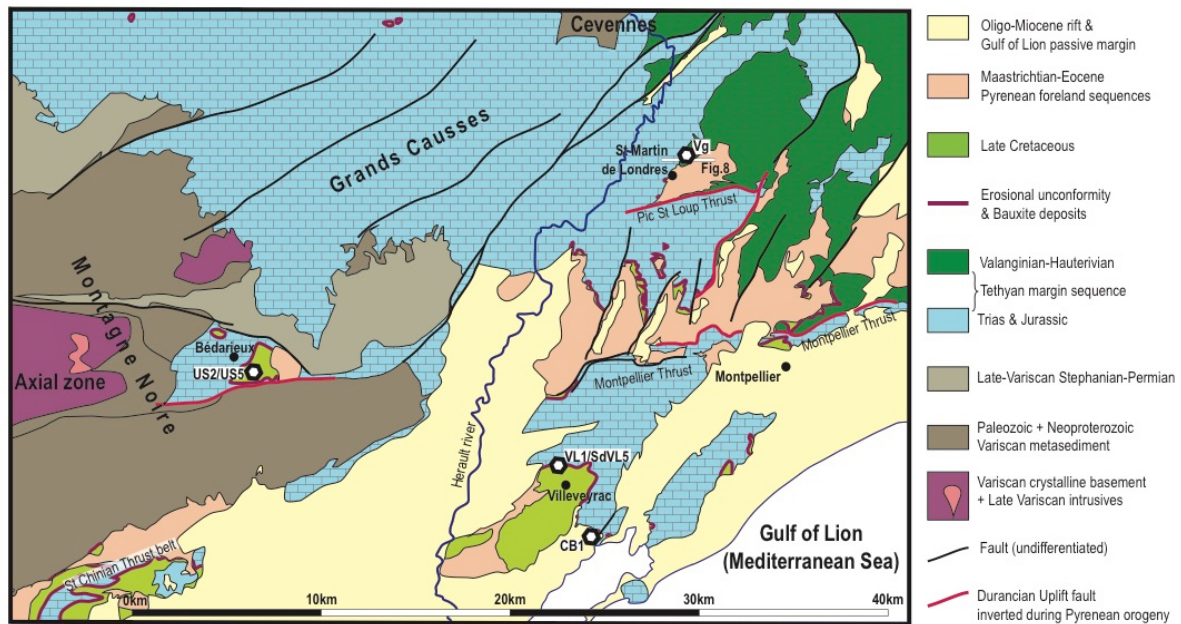


Fig. 3 : Simplified geological map of the study area (location on Fig. 1), showing the structural and stratigraphic context of the samples (hexagons). Synthetised from BRGM 1/250000 map of Montpellier (Alabouvette, B. *et al.*, 2001). The faults displaying evidence of activity during the Durancian uplift are underlined in red.

In the study area, the regional unconformity is outlined by bauxite deposits trapped inside lapiaz of the karstified Jurassic carbonates (Bardossy, G. & Combes, P.-J., 1999 ; Alabouvette, B. *et al.*, 2001). Remnants of bauxite are also found in the Jurassic plateaus of the Grands Causses (Fig. 3); they are covered with outliers of late Cretaceous shallow marine sediments (Bruxelles, L. *et al.*, 1999; Bruxelles, L., 2001). Other occurrences of bauxites are found in the Saint Chinian thrust belt (Fig. 3), where erosion due to the Durancian uplift reached down to the Lias limestone. In the sampled basins, bauxite deposits present a variety of lithofacies (Fig. 4). They consist of breccia and microconglomerates of reworked bauxite clasts (Fig. 4A, B) and bauxitic pisolithes (Fig. 4C), interbedded with clay, and silt levels displaying hydrodynamic figures. Some metre-scale blocks of stratified bauxite can be recognized in the deposits (Fig. 4D). These observations confirm an allochthonous origin (Demangeon, P., 1965 ; Combes, P.-J., 1969), although some additional post-depositional weathering may have occurred inside the deposit (Combes, P.-J., 1990). The origin of the allochthonous bauxite is still discussed. They were initially developed as a weathering cap over aluminous formations (crystalline rocks or clay-rich sediments), subsequently reworked and transported towards their present position. Their location, closely downstream of the Montagne Noire, as well as the presence of heavy minerals (especially millimetre-size tourmalines) suggest an origin from the Variscan basement (Combes, P.-J., 1990). Alternatively, clays and marls of a sedimentary cover can be weathered and can provide bauxitic materials, as demonstrated by in the Ariège-type bauxites (Fig. 1; (Combes, P.-J., 1969).

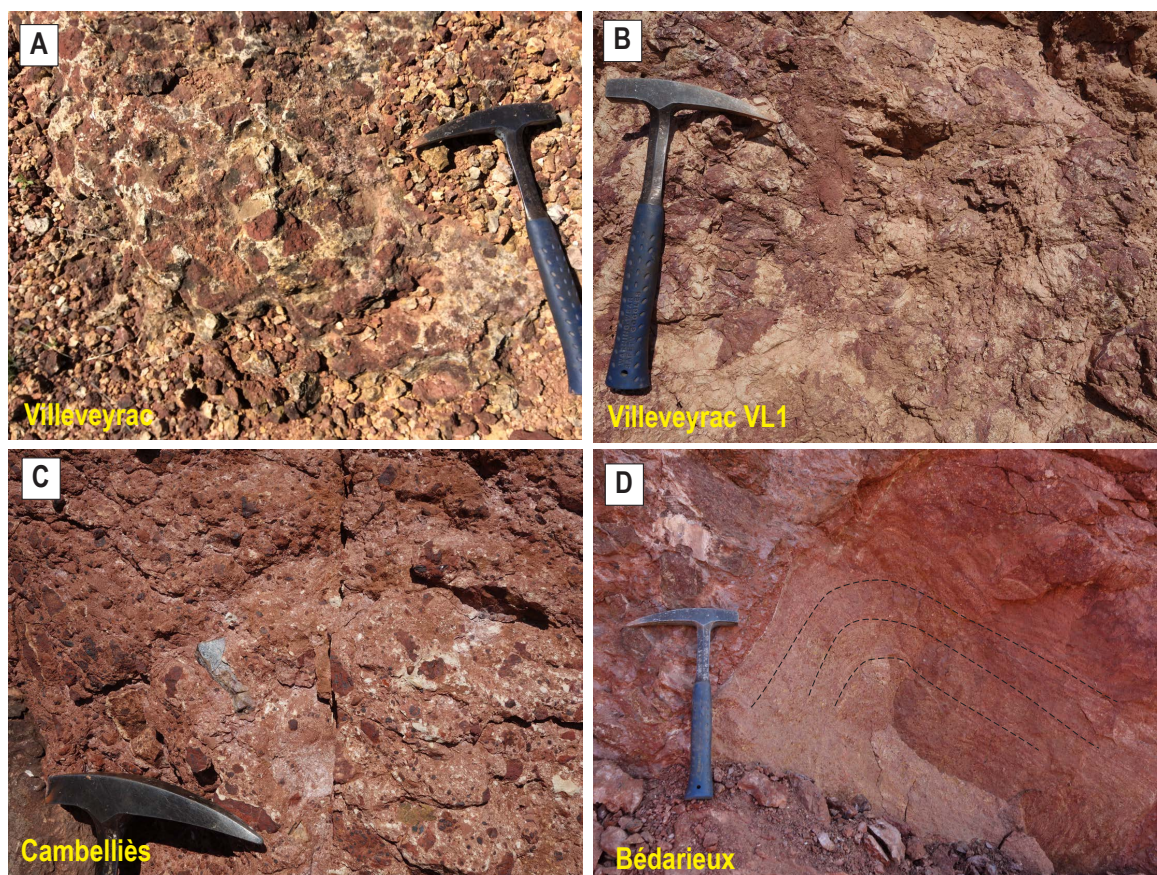


Fig. 4 : Different aspects of allochthonous bauxites. (A): Breccia of red bauxites elements in a white, low-Fe bauxitic matrix; (B): mass-flow deposit displaying bauxite elements of different origins, corresponding to VL1 sample ; (C): Bauxite breccia displaying: pisolithes, lateritic duricrust elements and grey Jurassic limestone; (D): mega breccia including a large clast of bauxite showing internal stratification, as evidence of poly-recycling.

The age of the bauxite deposits in southern France is poorly constrained as they do not contain any fossil (Lajoinie, J.-P. & Laville, P., 1979). The age of deposition is older than the karstified bedrock, and younger than the age of the sedimentary cover that caps the bauxite. The bauxitic cover ranges from Aptian in the Pyrenees (Combes, P.-J., 1990) to Eocene in Bédarieux (Combes, P.-J., 1969). The shortest hiatus is found in Provence, where bauxites are unconformable over Barremian and capped by Turonian (Lajoinie, J.-P. & Laville, P., 1979). In the study area (Fig. 3), the bauxites of the Villeveyrac basin are at the top of Late Jurassic karstified limestone and sealed by a glauconitic sandstone that yielded latest Albian (“Vraconian”) foraminifera as reported by M. J Fondécave in (Husson, E., 2013). Such a long hiatus implies an erosional episode prior to bauxite deposition. However, in Bédarieux, bauxite deposits consist of two superposed units interleaved with dolomite sandstones and shales, suggesting two bauxitic events: Berriasian and Aptian-Albian (Combes, P.-J., 1969). Evidence of the first event was supported by the identification of reworked pollen in the dolomite sandstone and shales (Combes, P.-J. *et al.*, 1973). This early (Berriasian) bauxitic event still remains unexplained within the regional framework.

3- Sample description

Three sample localities have been chosen from three bauxite basins where the aluminium ore has been, or still is, mined. They are located along a NW-SE transect (Fig.3) from close to the

Variscan basement (Bédarieux) to the present-day Mediterranean Sea (Cambelliès), as reported in Appendix 1. Sampling consisted of 2-3 kg of pisolithic-rich interval of allochthonous bauxite, filling lapiaz and sinkholes of the karstified Jurassic carbonates. All bauxite samples were collected in the filling of the lapiaz, close to the Jurassic wall, where detrital heavy minerals are more likely to accumulate. An additional sample was taken from the closest outcrop of Valanginian marls, in order to characterize the detrital zircon content of a potential Early Cretaceous sedimentary source for the bauxites (Fig. 3).

3-1. The Bédarieux outcrop (Samples US2, US5)

The Bédarieux karst bauxite basin is located to the east of the Montagne Noire gneiss dome (Fig. 3). The EW trending basin is bounded to the south by a normal fault separating the karstified Dogger sequences from the Variscan basement; the original fault was inverted during the Eocene Pyrenean orogeny (Bogdanoff, S. *et al.*, 1982). In the northern part of the basin, the ore bodies consist of two main sub-basins corresponding to distinct karstic pockets whose depth may exceed 70 m (Crepel, G., 2005), whereas in the southern part (where the samples were taken in the Uston district), it is constituted by a 5 to 10 m thick, rather continuous level, filling and covering karstified Bathonian dolomites. The bauxite are capped by continental Late Cretaceous (Lajoinie, J.-P. & Laville, P., 1979) and Early Eocene deposits (Combes, P.-J., 1969). No evidence of fault activity affecting the bauxite has been observed (Crepel, G., 2005). However, bauxite present slickensides concentrated along the karst wall, indicating post-deposition collapse movements, induced by karst processes (Combes, P.-J., 1969).

The bauxite deposits display a highly irregular base, due to the karstic nature of the footwall, inducing variable thickness (Fig. 5). At Uston, the sample locality, the interval is about 10m thick, comprising two bauxite levels (the “lower” and “upper bauxite”) separated by a 20 cm thick, sub-horizontal, bed of yellow detrital dolo-sandstone, which laterally passes to yellow marls. This “lower” bauxite is reputedly Berriasian in age (Combes, P.-J. *et al.*, 1973). Each bauxite unit presents an erosive base and onlaps onto the steeply dipping dolostone wall, resulting in the complete burial of the karst relief by bauxite deposits (Fig. 5). Two samples were taken from the lower and upper bauxites, US2 and US5 respectively.

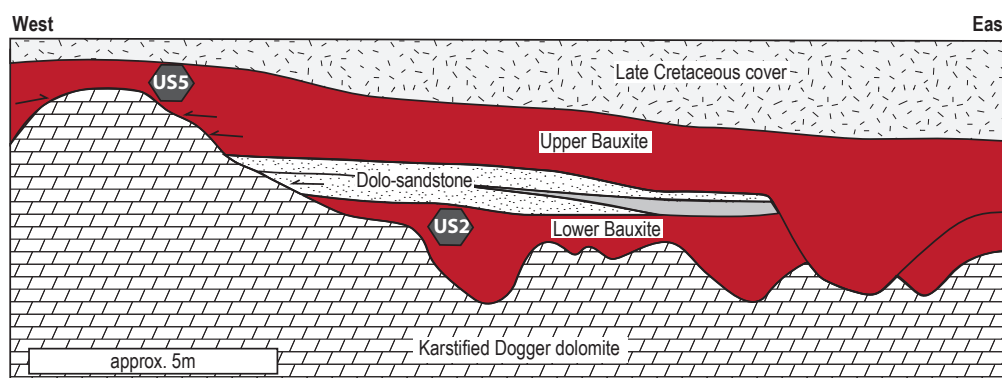


Fig. 5 : Cross section of the Bédarieux outcrop, showing the bauxite deposits (red) filling karstified dolomite and the location of samples US2 and US5. Two bauxite levels, separated by dolo-sandstones and marls, are onlapping the karst topography, and are covered by late Cretaceous conglomerates and sandstones. Note that the upper bauxite deposits are locally incised into the lower bauxite.

3-2. Villeveyrac outcrop (samples VL1, SdVL5)

The Villeveyrac basin corresponds to a syncline whose axis dips 10° southwest, involving the Mesozoic and Lower Eocene series (Fig. 3). In this basin, bauxites appear as a several metre-thick, continuous deposit over the entire basin. Mineralogical study of core-drill of the Villeveyrac bauxite and an analysis of the vertical and lateral distribution of Al_2O_3 , $(\text{FeO} + \text{Fe}_2\text{O}_3)$,

SiO₂ and TiO₂ across the basin can be found in (Marchand, E., 2019). The karstified Portlandian carbonate forms metre-deep juxtaposed depressions, making a general corrugated base for bauxite deposit. The lapiaz was formed by preferential carbonate dissolution along orthogonal joints (N060 and N150) (Marchand, E., 2019). The cover consists of marls, sandstone and lignite, indicating continental to very shallow marine environments, referred to Turonian to Senonian (Combes, P.-J., 1969). However, marine foraminifera identified by M.J. Fondécave, and reported in Husson (2013) indicate a Latest Albian ("Vraconian") age for the marine glauconitic sandstone (Fig. 6).

One sample was taken from the lower part of the bauxitic interval (VL1) thanks to a freshly cut section along an active extraction pit. Another sample was taken in the overlying marine glauconitic sandstone (SdVL5), which yielded the Latest Albian foraminifera (Fig. 6).

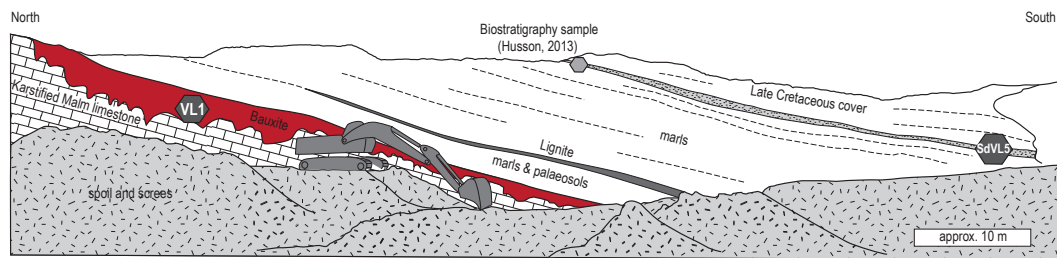


Fig. 6 : Section of the Villeveyrac bauxite open cast showing sampling localities. Bauxite deposits (sample VL1) are filling the karst, and are sealed by lacustrine to shallow marine sequences, especially a glauconitic sandstone (sample SdVL5), that yielded latest Albian foraminifera (Husson, E., 2013).

3-3. Cambelliès outcrop (Sample CB1)

The Cambelliès outcrop is the further away from the Montagne Noire and corresponds to a southward extension of the Villeveyrac basin. Prior to bauxite mining in the 1970', this basin was almost completely covered by Miocene marine sediment belonging to the Gulf of Lion margin (Fig. 3). The bauxite interval is mainly concentrated along a NE-trending normal fault affecting Upper Jurassic karstified limestone, and with evidence of syn-depositional activity (Fig. 7). The bauxite cap consists of Late Cretaceous lacustrine and palustrine limestones, marls and sandstones. The whole set is unconformably covered by the marine Early Miocene molasses, sandstones and marls, capped by a bioclastic limestone. One bauxite sample (CB1) was taken in the deep part of a lapiaz.

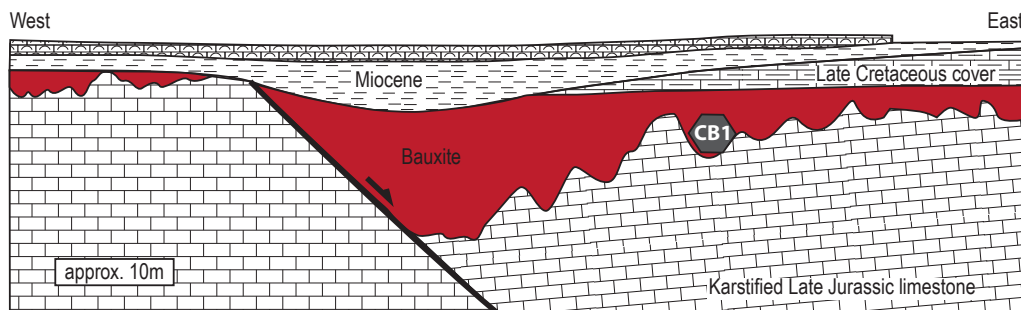


Fig 7 : Cross section of Cambelliès outcrop, where bauxite deposits (sample CB1) reveal a syn-depositional normal fault, sealed by Late Cretaceous lacustrine limestone and Miocene marls and bioclastic limestone.

3-4. St Martin de Londres Valanginian marls (Samples Vg)

Weathering and alteration of the clay content of marls may represent a possible source for the bauxites of Languedoc and Pyrenees (Combes, P.-J., 1990). The early Valanginian marls and marly limestones represent the closest marly formation to the Languedoc bauxites, both stratigraphically and geographically. Valanginian formations are lacking in the Bédarieux-Villeveyrac-Cambelliès areas as a result of erosion during the Durancian uplift. However, they are preserved in the St Martin de Londres synform, north of the Pic St Loup thrust, an inverted mid-Cretaceous normal fault (Fig. 3). The sampling outcrop is located 35km NE of Villeveyrac and 50km east of Bédarieux, respectively. It corresponds to lower Valanginian platform marls, that onlap onto the Late Jurassic and Berriasian, high-energy, shallow, marine limestone. They are unconformably covered by latest Cretaceous-Early Paleocene continental deposits (Fig. 8) (Philip, H. *et al.*, 1978). The > 60 My-long erosional hiatus (Valanginian - Latest Cretaceous) brackets the bauxite formation episode. Scarce bauxite deposits are trapped in the karst that affects the Late Jurassic only 2 km SW of the sampling area (Fig. 3).

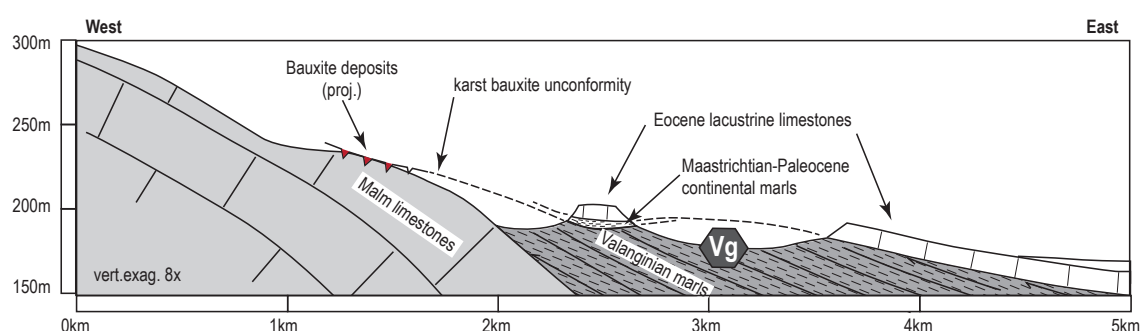


Fig 8 : Simplified section showing the stratigraphic and structural position of the sample taken in the Valanginian marls of St Martin de Londres (sample Vg).

4- Analytical method

Conventional heavy mineral separation methods (heavy liquids and magnetic separation) have been used to separate zircon grains from the studied samples. Extracted zircons were subsequently handpicked in alcohol under a binocular microscope. The grains were put in epoxy resin together with chips of the standard zircons GJ1 and G91500 (Wiedenbeck, M., 1995 ; Jackson, S. E. *et al.*, 2004), and polished to approximately half of their thickness to expose grain interior. Laser ablation ICP-MS analyses were performed with a spot size of 26µm, systematically aimed at the centre of the grain, using an excimer laser Compex 102 (LambdaPhysik, Geolas Platform) coupled to a single collector ICP-MS Element 2 XR from the AETE-ISO regional facility of the OSU OREME (Université de Montpellier) following the procedure outlined in earlier reports (e.g. (Van Achterbergh, E. *et al.*, 2001; Bosch, D. *et al.*, 2011; Bruguier, O. *et al.*, 2017). Ablation experiments were conducted under ultra-pure helium, which enhances sensitivity and reduces Pb-U fractionation (e.g. (Gunther, D. & A. Heinrich, C., 1999). The He gas stream and the particles from the samples were subsequently mixed with Ar shortly before entering the plasma. The laser was fired at an energy density of 12 J/cm² at a frequency of 3 Hz. Each analysis consisted of 15 s of gas blank followed by 45 s of signal acquisition. Before the analysis, the targeted zone was cleaned by 10 laser shots using a spot size bigger than the one used for the analyses (51 µm). Unknowns were bracketed by the sequential measurement of standards zircons which were analysed twice each five unknowns.

Data processing was performed with the GLITTER program (Van Achterbergh, E. *et al.*, 2001). Probability density plots and Concordia diagrams were drawn using the ISOPLOT program (Ludwig, 2003). In the discussion section, only concordant analyses were considered in order to establish sediment provenance. Concordance is defined by the ratio between the ages given by

the $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{206}\text{Pb}/^{238}\text{U}$ ratios. It is however noteworthy that the range of radiogenic $^{207}\text{Pb}/^{206}\text{Pb}$ throughout the Phanerozoic is too small to give a sensitive measure of the age, given the precision classically achieved with laser ablation ICP-MS analyses and low abundance of the ^{207}Pb (c. 20 times less than the ^{206}Pb). Thus for grains younger than 1 Ga, we used the $^{207}\text{Pb}/^{206}\text{Pb}$ ratio as the best age estimate (and a discordance threshold of $\pm 5\%$) whereas for grains younger than 1 Ga, the most reliable $^{206}\text{Pb}/^{238}\text{U}$ ratio was preferred. In addition, for reasons explained above, the discordance threshold was increased to $\pm 10\%$. Grains displaying a discordance level higher than 5 % (grains older than 1 Ga) or 10 % (grains younger than 1 Ga), were rejected from the density probability plots and not considered in the discussion.

5- Results

Results from the 6 samples (bauxites, cover sandstone and Valanginian marls) are provided as Analytical results table in Supporting information Appendix 2. They are presented as frequency histograms and probability density diagram (Fig. 9) and as $\pm 1\sigma$ in Concordia plot, in Supporting information Appendix 3. Ages in the text are quoted at the $\pm 2\sigma$ levels.

- Sample US2 (Bédarieux)

Among 148 analyses, 136 are concordant within errors and yield ages ranging from 113.5 ± 5.4 Ma to 2805 ± 45 Ma (2σ). The relative probability curve (Fig. 9) is dominated by a very sharp peak at c. 318 Ma representing 16 % of the analyses. Most grains however fall in the 500-750 Ma age range with age peaks at c. 500 Ma, c. 600 Ma and c. 675 Ma. Broad peaks appear at 950-1150 Ma, 1350-1450 Ma and 1750-1950 Ma with subordinate peaks at 2.1 Ga, 2.5 Ga and 2.65-2.80 Ga.

- Sample US5 (Bédarieux)

In this sample, 150 zircon grains have been analysed and 132 were found to be concordant. The age spectrum ranges from 118.0 Ma to 3549.5 Ma. Similarly to sample US2, the zircon age distribution is dominated by Variscan grains, with a sharp peak at c. 316 Ma (Fig. 9). Other grains have ages ranging from 450 to 850 Ma with a main distribution around 580 Ma. The remaining grains plot evenly between 1000 and 2800 Ma and it is difficult to recognize clear age peaks as was the case for sample US2. However, similarly to this sample, no detrital grains older than 2.9 Ga were detected, except for one grain which yields an Early Archean age of 3550 ± 26 Ma (2σ). The youngest grain analysed provides a maximum age for the deposition, which is 118.0 ± 8.0 Ma. This is similar to the youngest grains detected in sample US2.

- Sample VL1 (Villeveyrac)

146 grains were analysed among which 136 are concordant, with ages ranging from 285 Ma to 2900 Ma (Fig. 9). The age spectrum is characterized by a multimodal distribution and is dominated by Variscan grains with sharp age peaks at c. 295 Ma and c. 360 Ma. Phanerozoic grains also include Ordovician (c. 475 Ma) and Silurian (c. 420 Ma) zircons. The next age peak includes Cambrian to Neoproterozoic grains with a maximum distribution at c. 550 Ma. Grains older than 1000 Ma, define age peaks at around 1000 Ma, 1800 Ma and 2700 Ma and represent more than 37 % of the whole dataset.

- Sample CB1 (Cambelliès)

Among 148 analyses, 140 are concordant ranging from 273.9 Ma to 3150.5 Ma (Fig. 9). The Variscan ages correspond to a large peak at c. 330 Ma. The two subordinate peaks are: 576 Ma and 414 Ma. Ages older than 1000 Ma represent 30 % of the sample and characterize two stretched out peaks around 1100 Ma and 1700 Ma. A broad peak is also present at around 2700 Ma and indicates the occurrence of late Archean grains.

- Sample SdVL5 (Villeveyrac)

In this sample, 78 zircon grains were analysed and 64 were found to be concordant ranging from 299.3 Ma to 2916.5 Ma (Fig. 9). The youngest peaks (298 Ma and 438 Ma) are narrower whereas towards older ages, peaks splay out. The main age peak is Neoproterozoic (630 Ma) and represents 19% of the whole data set, whereas the Variscan age peak corresponds to 3%. It is noteworthy that 50% of the zircon grains are represented by the three subordinate peaks around 1100 Ma, 1700 Ma and 2700 Ma.

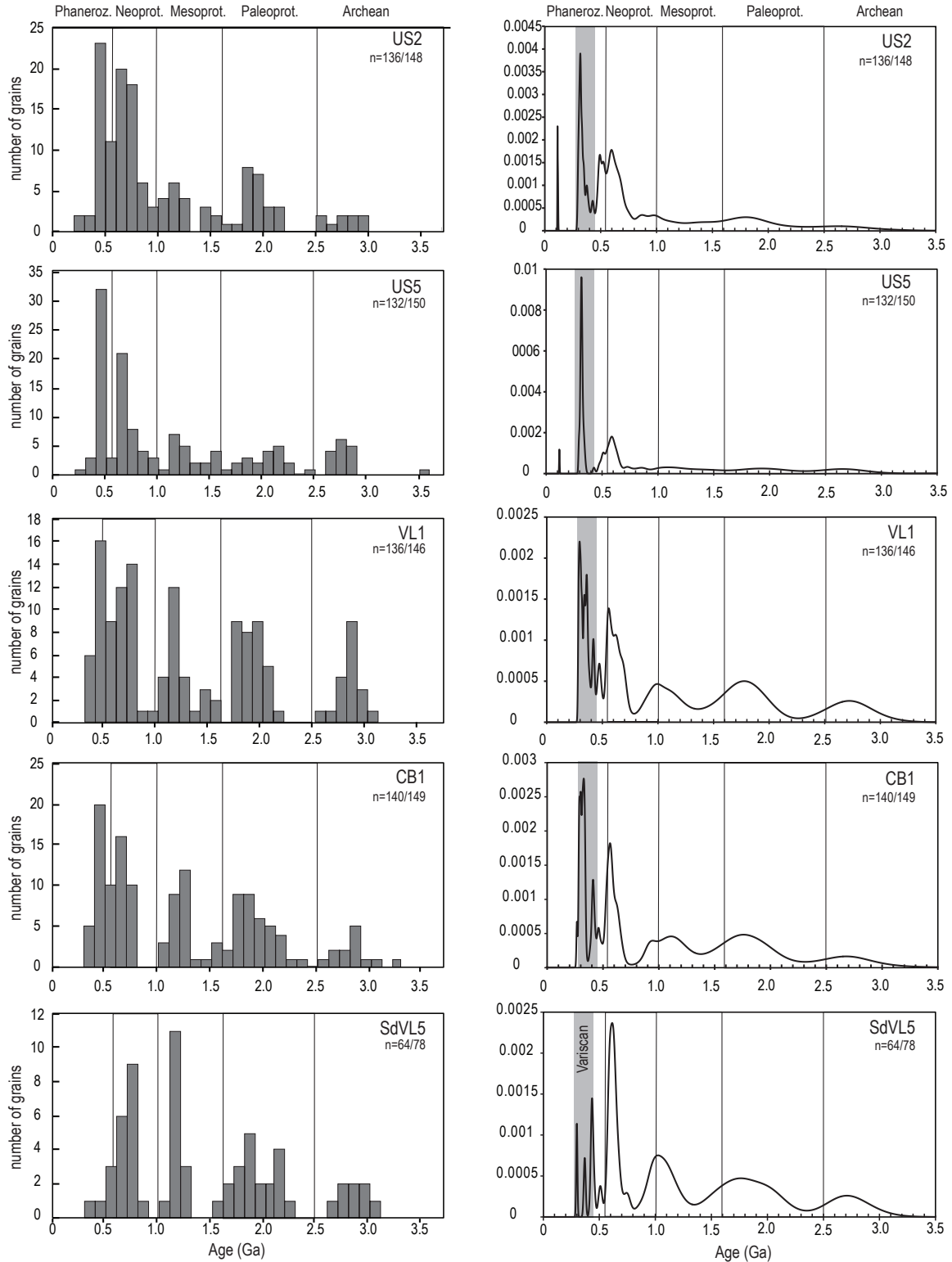


Fig. 9 : Results of U-Pb LA-ICP-MS analyses of detrital zircons from the bauxite basins. Left column: age distribution of concordant analyses (histogram binwidth : 100 Myrs). Right column : relative

probability curve with concordant analyses; grey shading indicate the Variscan cycle interval (450-290 Ma). US2, US5, VL1 and CB1 are bauxite samples; SdVL5 corresponds to the glauconitic sandstone cover of the VL1 bauxite.

- Sample Vg (St Martin de Londres)

As expected, detrital zircons were very scarce in the marls: only 11 zircons were found in the > 20 kg sample. All grains are concordant, ranging from 133 to 1871 Ma, but such limited number of results must be interpreted with caution (Fig. 10). Nevertheless, the youngest grain (133±3 Ma), although slightly discordant, corresponds to the stratigraphic age of the sampled formation (Valanginian) ; it could be related to volcanism, although no magmatic event of this age is recorded in the area. In spite of the very small number of grains, which does not allow to distinguish peaks, it can be noted : three Variscan ages (298, 342, 352 Ma), three Early Paleozoic ages (440, 475, 553 Ma) , two Neoproterozoic ages (730 & 1009 Ma) and two late Paleoproterozoic age (1852 & 1871 Ma). Except for the Valanginian date, all these age groups are found in the bauxite samples, and no additional date group is present in the sample.

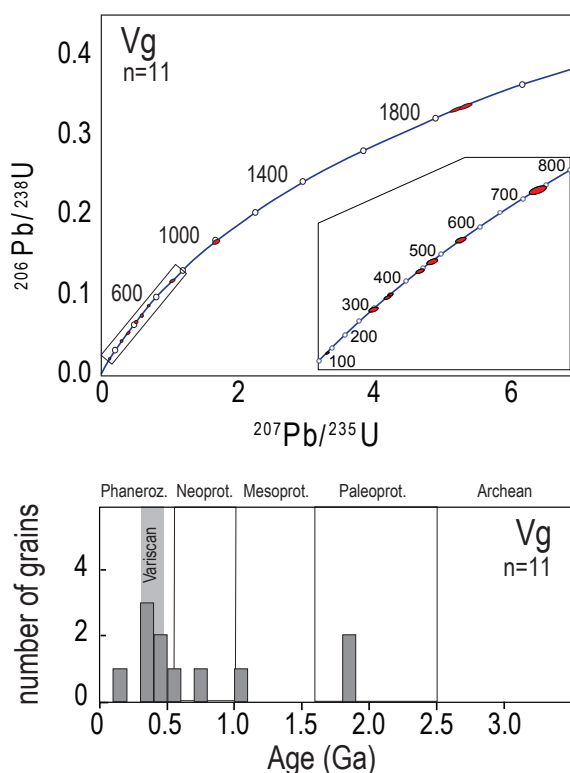


Fig. 10 : Analytical results of U-Pb LA-ICP-MS analyses of detrital zircons from the Valanginian marls from St Martin de Londres. Concordia diagram (Inset: 100-800 Ma) and histogram (binwidth : 100 Myrs).

6- Interpretation of detrital zircons results

6-1. Age of the Languedoc bauxite deposits

The youngest detrital zircon grains were found in the two Bédarieux samples (US2 and US5) and display mid-Cretaceous ages of 113.5 ± 5.4 Ma and 117.3 ± 7.8 Ma in the US2 sample and 118.0 ± 8.0 Ma in the US5 sample. The three ages overlap within errors and can be pooled to define a mean age of 115.5 ± 3.8 Ma (MSWD = 0.6 ; n = 3). This age corresponds to a major pulse in volcanic activity in the Tethyan realm (Charbonnier, G. & Follmi, K. B., 2017; Sabatino, N. *et al.*, 2018). After LA-ICP-MS analyses, the grains were subsequently observed using a binocular microscope. Observations revealed sharp edges for two grains (ml32 – mc12, see Supporting information Table 2) consistent with a short distance transport, which is in agreement with a volcanic, potentially airborne, origin. A precise location of the potential source of these zircons is

out of the scope of this contribution. However, it is worth noting that magmatism of mid-Cretaceous age is described in the Pyrenees (Montigny, R. *et al.*, 1986) and that nepheline syenites interbedded within Aptian sediments occur in the eastern Pyrenees (Azambre, B., 1970; Berger, G. M., 1982; Berger, G. M. *et al.*, 1997), some 90 km away from the Bédarieux basin. This area, and the volcanic products associated with the nepheline syenites, might represent the source of the mid-Cretaceous zircons. An alternative source, although located further west, could be the alkaline volcanics dated to the late Albian, found in the North Pyrenean Basins and the Basque Basin (Ubide, T. *et al.*, 2014). Finally, one could also hypothesize the existence of a closer source, now removed by Pyrenean deformation and erosion, or presently buried beneath younger sequences. Our new results allow to better constrain the age of bauxite deposition. The Upper Aptian – lower Albian age of 115.5 ± 3.8 Ma found in the volcanic zircons, embedded in the bauxites deposits, is taken as a oldest possible age for bauxite deposition. Bauxites are therefore younger than 115.5 Ma (Late Aptian) and older than Late Albian cover of the bauxite, according to the biostratigraphic results (Husson, E., 2013). The bauxite deposits of Languedoc can therefore be dated to the Early to mid-Albian. These new results allow to propose that the period for bauxite deposition in southern France can be bracketed between 115.5 Ma and the late Albian, “Vraconian” (Husson, E., 2013), thus restraining the bauxite formation, reworking and trapping in the Languedoc basins to a *circa* 10 My long interval. The identification of these mid-Cretaceous zircon grains in both the upper and lower bauxites in Bédarieux, clearly points to a relatively short bauxite deposition event. Our results also rule out the hypothesis of a Berriasian age for the lower bauxite (Combes, P.-J. *et al.*, 1973). On the basis of these results, the microfossils and pollen found in the marls between the lower and upper bauxites in the Bédarieux basin must be considered as reworked material, deposited along with the bauxites. Reworking of the wall sequences is also suggested by the occurrence of dolomite sandstone material, derived from erosion and reworking of the karst pinnacles, which is interbedded within the bauxite deposits (Fig. 5)

6-2 Two types of source for the bauxite

The source for the studied allochthonous bauxites must be understood as reworking of weathered and bauxitized profiles, which were developed on top of an aluminous-rich substrate: crystalline/metamorphic basement rocks, or non-metamorphic shale/marl sediments (Tardy, Y. & Roquin, C., 1998). Bauxite is a weak material, which cannot be transported over long distance. This is supported by the mass transport process displayed by the allochthonous bauxite (Marchand, E., 2019), which preserves tens of centimeters-large clasts of the original bauxite (see Fig. 4). Zircon U-Pb results from the 4 bauxite samples show a spectrum of age peaks ranging from 113 Ma to 3.5 Ga (Fig. 9). Five age clusters or peaks are distinguished in the four bauxite samples, despite varying proportions: centered on 350 Ma, on 600 Ma, from 1000 to 1200 Ma, between 1600 to 1900 Ma and centered on 2700 Ma, respectively. The diversity of dates found, as well as the lack of identified local magmatic sources of Mesoproterozoic to Archean plutons, indicate that part of the zircons dated are either inherited zircons or derived from recycled (meta-) sedimentary sources.

The weathered profiles of the adjacent Variscan basement of the Montagne Noire, is one likely source for the detrital component of the bauxites. Gneisses occur in the Axial Zone of the Montagne Noire (Fig. 3), with an average age of 450 Ma for the protoliths (Roger, F. *et al.*, 2004 ; Pitra, P. *et al.*, 2012). The late Variscan evolution was characterized by the emplacement of late granitoids, and their volcanic equivalents, during the 290-320 Ma period (Bruguier, O. *et al.*, 1998; Bruguier, O. *et al.*, 2003 ; Poujol, M. *et al.*, 2017 ; Trap, P. *et al.*, 2017), which can be the source of late Paleozoic zircons in the bauxites. Furthermore, detrital zircons from the pre-Variscan Ediacaran-Cambrian to Devonian metasediments, found in the recumbent folds around the Montagne Noire axial zone, yielded a variety of ages, with predominance of Late Proterozoic and Cambrian to Ordovician zircons (Gebauer, D. *et al.*, 1989 ; Lin, W. *et al.*, 2016 ; Padel, M. *et al.*, 2017). In addition to these zircon populations, Late Carboniferous syn-orogenic formations include an additional, highly represented, group of Early Carboniferous ages (360-340Ma) (Lin,

W. *et al.*, 2016 ; Martínez, F. J. *et al.*, 2016), which are also found in the bauxites. The proportion of Variscan zircons (360 to 290 My) decreases away from the Montagne Noire. Such grains represent a quarter of the Bédarioux samples (15 km from the axial zone and 1 km from the Paleozoic Variscan recumbent folds), but only 14 % in the Cambelliès sample (50 km SE of the Montagne Noire). These observations are therefore consistent with one local source of the allochthonous bauxite, originated in the Montagne Noire and corresponding to the basement structural high affected by the Variscan orogeny, and including inherited protoliths of the European continental crust (Melleton, J. *et al.*, 2010).

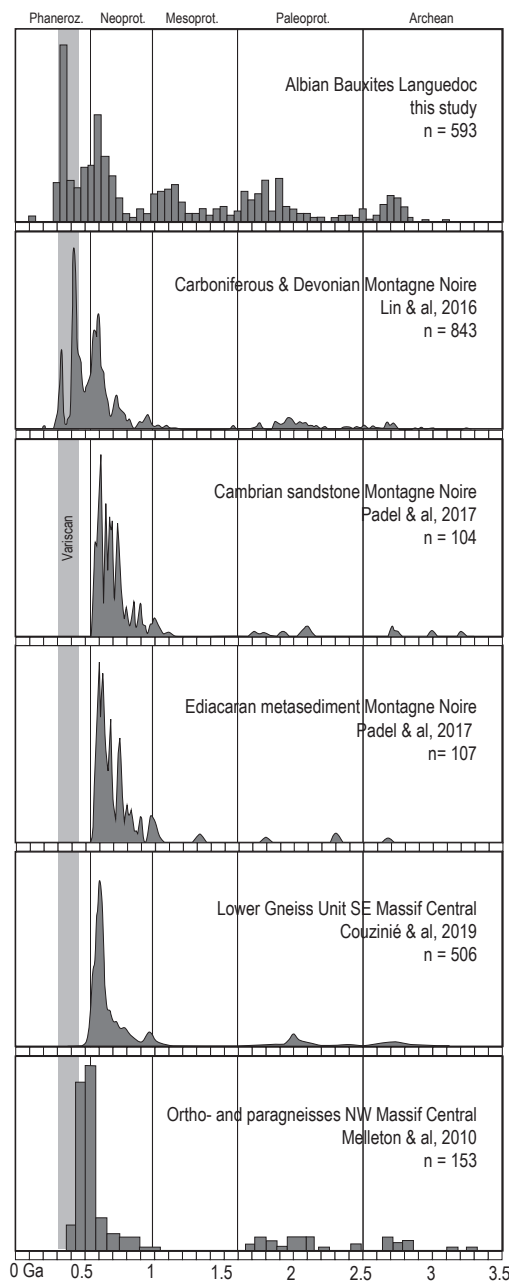


Fig. 11 : Comparison of our U-Pb LA-ICP-MS analyses of detrital zircons from bauxites (top panel histogram binwidth = 50Myrs) with detrital zircon ages from the Variscan basement (metasediment from the Montagne Noire and gneisses from the Massif Central). Histograms and kernel density diagrams are reproduced from previous studies, they include different vertical scales. The age spectrum from the bauxites (top panel) displays large proportions of Mesoproterozoic, Paleoproterozoic and Archean ages, which cannot be sourced from the Variscan basement (lower panels) as these rocks contain very little proportions of grains with such ages. Such comparison thus suggests an additional, sedimentary, source. Geological eras are plotted to ease correlations (see discussion in text).

Comparing the ages of the detrital zircons from the bauxite with the detrital zircons from (meta-) sediments and zircons from the basement rocks of the Montagne Noire (Fig. 11), points to significant differences. Bauxite yield zircons covering almost all the age range from late Variscan to Archean. By contrast, protoliths from the southern Massif Central, shows widespread Neoproterozoic ages, but Meso- and Paleoproterozoic ages as well as Archean ages, are scarce, either in the axial zone or in the recumbent fold of the Montagne Noire (Lin, W. *et al.*, 2016;

Padel, M. *et al.*, 2017). Old zircons may be present as isolated, inherited grains in the paragneisses, from the more distant eastern (Couzinié, S. *et al.*, 2019), or western Massif Central (Melleton *et al.*, 2010). Therefore, the significant clusters of late Mesoproterozoic, Paleoproterozoic and Archean dates, observed in the bauxites (18%, 21% and 6%, respectively) strongly differ from age spectra found in U-Pb studies from the Variscan basement.

This suggests a second, distinct, contributor to the allochthonous bauxite deposits. This source is recorded in the three studied basins (although more diluted in the proximal Bédarieux samples, due to the dominant Variscan basement source), which points to a regionally distributed, generalized, source. In addition, the wide and continuous range of ages found in the bauxite zircons suggests multiple mixing and recycling, in agreement with reworking of a sedimentary source. Indeed, recycling of (meta-)sediments into younger sequences promotes mixing of heavy minerals derived from diverse sources, potentially distant and/or now completely removed by erosion. The original provenance of Mesoproterozoic, Paleoproterozoic and Archean zircons, probably poly-recycled, is questionable. However, the source for the older zircons could be sought north of the study area (Scotese, C. R., 2014), across the northwest European platform, in North America and Scandinavia (Gee, D. G. & Stephenson, R. A., 2006; Whitmeyer, S. & Karlstrom, K. E., 2007; St-Onge, M. *et al.*, 2009).

Therefore, the age frequency distribution of detrital zircons from the four bauxites samples could be accounted for by mixing of two sources:

A generalized sedimentary source that equally fed all investigated basins, characterized by 4 significant date-clusters (Late Neoproterozoic, Late Mesoproterozoic, Paleoproterozoic, and Neoproterozoic).

A localised source from the Variscan basement of the Montagne Noire, whose signature includes Late Variscan and Late Neoproterozoic ages, which rapidly decreases southeastwards.

6-3. Montagne Noire progressive uplift

The two superposed Bédarieux samples present similar spectra with varying proportions. The lower bauxite (US2) displays dominant Variscan and Early Paleozoic ages, including a late Variscan peak at around 300 Ma, which represents 17% of the dated zircons. Early Paleozoic to Late Neoproterozoic zircons are interpreted as derived from the metasediments of the Variscan allochthon (Gebauer, D. *et al.*, 1989 ; Lin, W. *et al.*, 2016) and from the protolith of the Axial Zone gneisses.

The upper bauxite (US5) displays a more pronounced peak at around 300 Ma, reaching 26.5% of the zircons in the sample, significantly more prevalent than the late Neoproterozoic peak. This temporal evolution could be interpreted as a change in catchment areas. Alternatively, it may reflect increasing erosion, which first removed large parts of the metasediments and basement gneisses, and later exhumed more late Variscan leucogranites, which intrude the Axial Zone gneisses (Poilvet, J.-C. *et al.*, 2011 ; Roger, F. *et al.*, 2015 ; Poujol, M. *et al.*, 2017). The observed stratigraphic bauxite sequences superposition in Bédarieux basin would therefore correspond to the reverse profile in the source area due to progressive denudation.

6-4. Late Albian bauxite cover

In the Villeveyrac bauxite basin, the bauxite deposits are topped by glauconitic sandstone (SdVL5) containing foraminifera, which indicates a marine environment (Husson, E., 2013). Detrital zircon grains have revealed dates groups similar to the bauxite samples in Villeveyrac and Cambelliès, except for the reduced number of Variscan ages (only 3% of zircons grains). This suggests that i) Late Albian sandstone cover was derived from the same source as the bauxites and ii) the supply from the local Variscan source decreased with time. The later indicates that the Montagne Noire, which had been actively uplifting during bauxite deposition

(Early to Middle Albian), was not being actively eroded by that time. In turn, this implies that tectonics and uplift of the hinterland had ceased by latest Albian.

7- Geodynamic implications

7-1. A Valanginian sedimentary cover on the southern Massif Central?

Based on the results presented here, tracing the sources of the bauxites in Languedoc leads us to argue for a regional, homogeneous, sedimentary cover, extending across the present outcrops of Variscan basement and Mesozoic Tethyan carbonate platform. The age of the sedimentary cover that acted as one of the two main sources for the bauxites and which later, provided detrital zircons to the latest Albian marine sandstone, can be discussed taking into account the stratigraphic relationships. The bauxitized sediments must be older than the allochthonous bauxite (Albian), and younger than the wall of the bauxite deposits, i.e. Dogger in Bédarieux and Malm in Villeveyrac and Cambelliès. The Valanginian marls and marly limestones interval is a good candidate as it contains significant amount of aluminous-rich clay minerals, prone to provide bauxites when altered (Combes, P.-J., 1969 ; Bardossy, G., 1982). The Valanginian marls sampled in St Martin de Londres contain very few detrital zircons (11 grains in circa 20 kg of sediment). However, carbonate dissolution and weathering of the clay content, leading to bauxite formation, do concentrate heavy minerals in the laterite. Zircon counting in our bauxite samples (about 100 grains in 1 kg) suggests that bauxitization of Valanginian marls would correspond to a concentration of heavy minerals by two orders of magnitude. We therefore hypothesize that weathering of the Valanginian sequence, which was originally several hundred metres thick (Gayte, D., 1984); (Amouroux, G., 2003), has provided a significant part of the material for bauxite generation. Our U-Pb dating of single detrital zircons from the Valanginian, although hampered by the small size of the sample, does not contradict this hypothesis.

Apatite fission-track analyses in the Variscan basement of southern France (Barbarand, J. *et al.*, 2001; Peyaud, J. B. *et al.*, 2005); (Olivetti, V. *et al.*, 2020) suggest one to two-kilometre amplitude denudation during the mid-Cretaceous. The exhumed post-Variscan peneplain, which displays Trias to Lias remnants indicates that the mid-Cretaceous erosion did not remove thick sections of the crystalline basement. Instead, denudation must have been done at the expense of a sedimentary cover, now removed, which was younger than Lias (preserved on the basement) and older than the age of denudation, i.e. Early Cretaceous in age (Barbarand, J. *et al.*, 2001; Séranne, M. *et al.*, 2002). Indeed, residual bauxite trapped in karstic features in the Jurassic high plateaus of the “Grands Causses” (Fig. 3) (Bruxelles, L., 2001), suggests that this sedimentary cover extended northward, across the southern French Massif Central (Thiry, M. *et al.*, 2006).

7-2. Mid-Cretaceous differential vertical movements

Modeling results of apatite fission-track thermochronology from the Variscan basement and Permian basins located north of the study area shows an episode of cooling between 110 and 100 Ma, which is interpreted to be the result of basement uplift and erosion (Barbarand, J. *et al.*, 2001; Séranne, M. *et al.*, 2002; Peyaud, J. B. *et al.*, 2005); (Pfeifer, L. S. *et al.*, 2018) (Olivetti, V. *et al.*, 2020) (). The best age constraint on bauxite deposition (Early to Middle Albian) therefore correlates with uplift and denudation of the Variscan basement and its sedimentary cover, in the hinterland. This uplift event was also contemporaneous with basin subsidence and deposition of marine sequences further south (Fig. 12). In southern Provence, marine Albian sequences were deposited in WSW-ENE elongated basins, resulting from in a NE-SW extension (Philip, J. *et al.*, 1987). Offshore Marseille, seismic reflection displays depocentres (> 1 km-thick), controlled by EW normal faults of undifferentiated Aptian-Albian age (Fournier, F. *et al.*, 2016). In the Corbières, southwest of the study area, strong thickness variation (0 – 1200 m), detrital fluxes within the overall carbonate shallow marine sediment, and rapid facies changes within the Albian sequence, suggest syntectonic deposits in rapidly subsiding basins (Mongin, D. &

Peybernes, B., 1980). In the eastern Pyrenees, half-grabens record Aptian syntectonic shallow marine sedimentation, followed by an Albian deepening, related to lithospheric extension (Chelalou, R., 2015; Chelalou, R. *et al.*, 2016). In the central Pyrenean domain, Aptian to early Albian marls and carbonates accumulated in moderately subsiding platform (several hundred metres accommodation), gave way in mid-Albian to early Cenomanian times to deposition of syn-tectonic, deep environment “Flysch noir” in depocentres, locally more than 3 km thick (Peybernes, B., 1982; Souquet, P. *et al.*, 1985). They were deposited in fast subsiding fault-controlled basins, along an EW trend with evidence of left-lateral displacement (Debroas, E.-J., 1990).

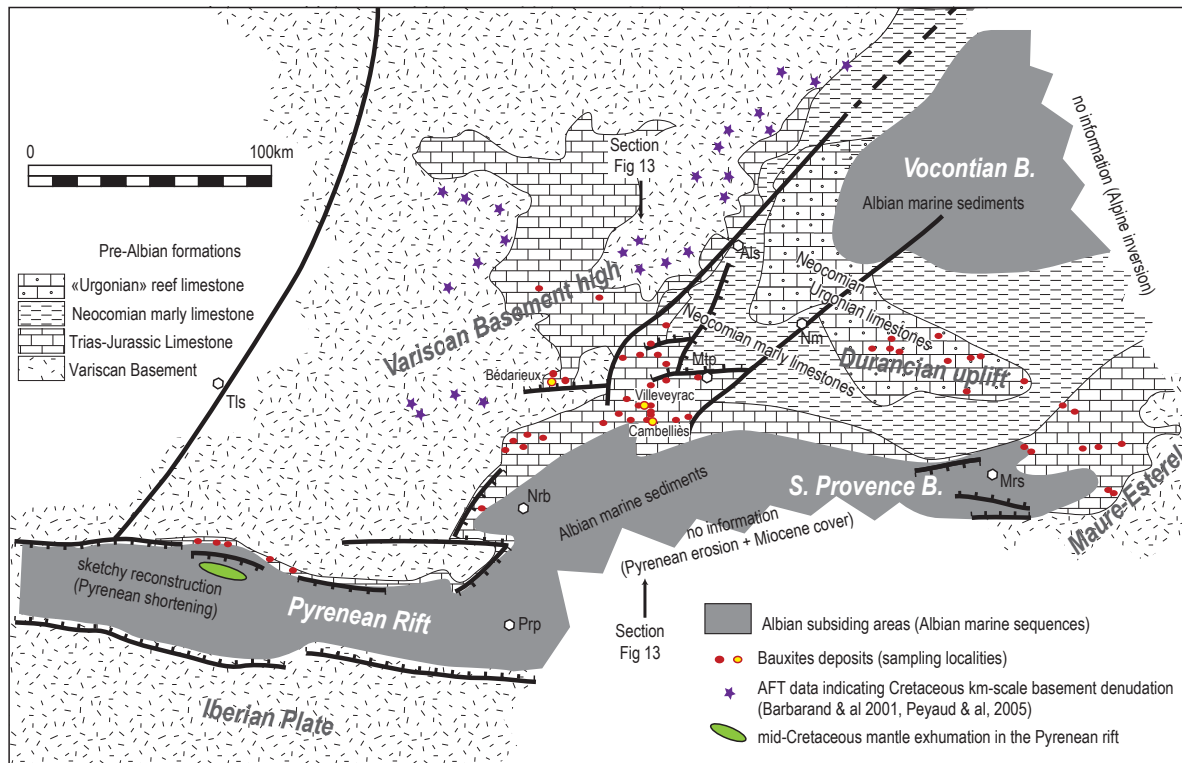


Fig. 12 : Interpreted structural and paleogeographic map during mid-Cretaceous time in southern France. Pre-Albian formations are represented in white with different overlays, while subsiding Albian basins are represented in dark grey. In the south, the rift extends in the Pyrenean Rift and in the south-Provence Basin. To the north, the Vocontian basin constitutes another distinct depocentre. Note that the Albian rift is much wider than the present distribution of Albian outcrops, due to later Pyrenean NS shortening. Unknown boundaries (covered by younger sequences) are represented as broken line. The “Durancian Uplift” lies between the two subsiding areas. Bauxites (red dots) are deposited on the karstified pre-Albian limestones of the uplifted area (Lajoinie, J.-P. & Laville, P., 1979; Combes, P.-J., 1990). Note that the marly Neocomian (Valanginian) cover that protected Jurassic from karstification does not bear bauxite deposits, whereas the overlying Neocomian limestone (“Urgonian” facies), exposed in Provence, has been karstified and contains bauxite deposits. The Variscan basement structural high provided Apatite Fission Track results (purple stars) consistent with an uplift-related denudation at around 100 My (Barbarand, J. *et al.*, 2001; Peyaud, J. B. *et al.*, 2005). Position of the main cities are given for reference: Als: Alès, Mrs: Marseille, Mtp: Montpellier, Nm: Nîmes, Nrb: Narbonne, Prp: Perpignan, Tls: Toulouse.

The Early to Middle Albian Languedoc bauxite deposition occurred along a hinge zone between the uplifting southern French Massif Central and onshore Provence (the « Durancian uplift») on the northern side, and subsiding offshore Provence-Corbières-Pyrenees, on the southern side. Albian bauxites were deposited and preserved at the base-level in the coastal plain. The closest marine sedimentation correlative to Languedoc bauxites is found 70 km southwestward, near

Narbonne and in boreholes near Béziers (50 km). The original paleogeographical setting has been disturbed due to later Pyrenean / Alpine thrusting and erosion, as well as Oligocene rifting and subsidence. Indeed, the paleogeographical map of Figure 12 is speculative in the southeast and east due to latter intense deformation (NW Mediterranean back-arc extension and Cenozoic Alps orogeny).

One outstanding question is the denudation by karstification that affected the Late Jurassic limestones, locally down to Dogger, as in Bédarieux. These karstified formations are unconformably covered by bauxite, which seems to contradict the presence of an Early Cretaceous sedimentary cover as a source for the bauxite. Structural framework analysis of the study area shows that denudation of the early Cretaceous cover was controlled by block faulting prior to bauxite allochthonous deposition (Fig. 3). We suggest that during the Aptian, 1) the northern part of the area (Montagne Noire to Cevennes) was affected by a regional uplift expressed by long-wavelength doming, where the cover was partly eroded, and the remnant was weathered in-situ, while 2) the south (study area) was tectonically active, affected by short-wavelength block faulting and differential vertical movements, which favored the total erosion of the Neocomian cover and karstification of the Jurassic limestone. The remaining marly Neocomian cover provided the protore for bauxite, which was later deposited on the karstified Jurassic limestone in the coastal area. Karstification of the Jurassic limestone and bauxitisation of the Valanginian marls and basement rocks were favoured by an Early Cretaceous tropical warm ($\geq 25^{\circ}\text{C}$) and humid ($\geq 1600\text{mm}$ precipitation) climate (Marchand, E., 2019; Chanvry, E. *et al.*, submitted).

7.3 Mid-Cretaceous geodynamic framework

In the Pyrenees, Aptian-Albian basin subsidence is related to an episode of rifting leading to lithospheric mantle denudation (Lagabriele, Y. & Bodinier, J.-L., 2008; Lagabriele, Y. *et al.*, 2010), and westward, to the opening of the Bay of Biscay (Jammes, S. *et al.*, 2009). Plate reconstruction models of Iberia vs Eurasia motion, relying on Atlantic magnetic anomalies, are conflicting (Olivet, J. L., 1996 ; Sibuet, J.-C. *et al.*, 2004; Vissers, R. L. M. & Meijer, P. T., 2012 ; Barnett-Moore, N. *et al.*, 2016 ; Van Hinsbergen, D. J. *et al.*, 2017). Eastward extension of the Cretaceous Bay of Biscay - Pyrenean Rift, where no oceanic magnetic anomalies are preserved, is even less resolved. In the study area, mid-Cretaceous tectonics is poorly understood. Few NE-trending faults display a moderate normal offset, of which Figure 7 is an example. In southern Provence, mid-Cretaceous tilting and normal faulting have been documented in relation to bauxite deposits (Guyonnet-Benaize, C. *et al.*, 2010), although it is not clear whether such upper crustal deformation accommodates lithospheric doming or results from a restraining bent in a left-lateral E-trending shearing. There is no onshore evidence of an eastward development of the Pyrenean Albian rifting. It could possibly be found offshore in the continuity of the North Pyrenean Fault, masked by the Pyrenean orogeny and Oligocene rifting (Guennoc, P. *et al.*, 1994; Mauffret, A. *et al.*, 1995; Séranne, M. *et al.*, 1995).

Attempts to link Iberia movement to the Alpine province paleogeography, through the present study area, are scarce and of low resolution for the scope of our local study (e.g. (Stampfli, G. M. & Kozur, H. W., 2006; Handy, M. R. *et al.*, 2010; Schettino, A. & Turco, E., 2011 ; Tavani, S. *et al.*, 2018). All these kinematic models, however, accommodate the anticlockwise rotation and eastward translation of the Iberian plate and related continental blocks (Alkapec terrane (Bouillin, J. P. *et al.*, 1986) with left-lateral transcurrent movements with respect to the south European plate. We hypothesize that differential uplift (Durancian isthmus) and subsidence (Pyrenean Rift, South-Provence and Vototian basins), which affected the south European margin during Albian, results from wrenching along NE-trending transform faults, in association with the anticlockwise rotation of Iberia-Alkapec.

Figure 13 presents the tectono-stratigraphic evolution of the upper crust in southern France. First, the Valanginian through to Hauterivian period records generalized subsidence of the entire region (Stampfli, G. M. *et al.*, 2002) (Fig. 13A), allowing deposition of about 1 km thick

marls and marly limestone (Dubois, P. & Delfaud, J., 1989; Bonijoly, D. *et al.*, 1996). The clay and silt content corresponds to the detrital products of several weathering episodes and successive erosional - depositional events, which allowed the multiple reworking of zircon grains, some of them as old as 3.5 Ga.

Barremian and Aptian series are not preserved in the study area, but neighbouring outcrops allow to document deposition of a regressive sequence characterized by high-energy reefal limestones (« Urgonian » facies) (Tendil, A. J.-B. *et al.*, 2018), passing eastwards to deeper environment, while the continental hinterland, corresponds to the Variscan basement high (Arnaud-Vanneau, A. *et al.*, 1979). Barremian-Aptian is interpreted as a period of moderate uplift of the hinterland (i.e. the study area), leading to emersion and continental weathering of the sedimentary cover (Fig. 13B). Further south, notably in the study area, block faulting preserved part of the Valanginian formation in the hanging-wall while in the footwall, Malm limestones were uplifted, denuded and karstified (Husson, E., 2013). Further south, South Provence and the future Pyrenees underwent rifting, subsidence and sediment deposition (Fournier, F. *et al.*, 2016).

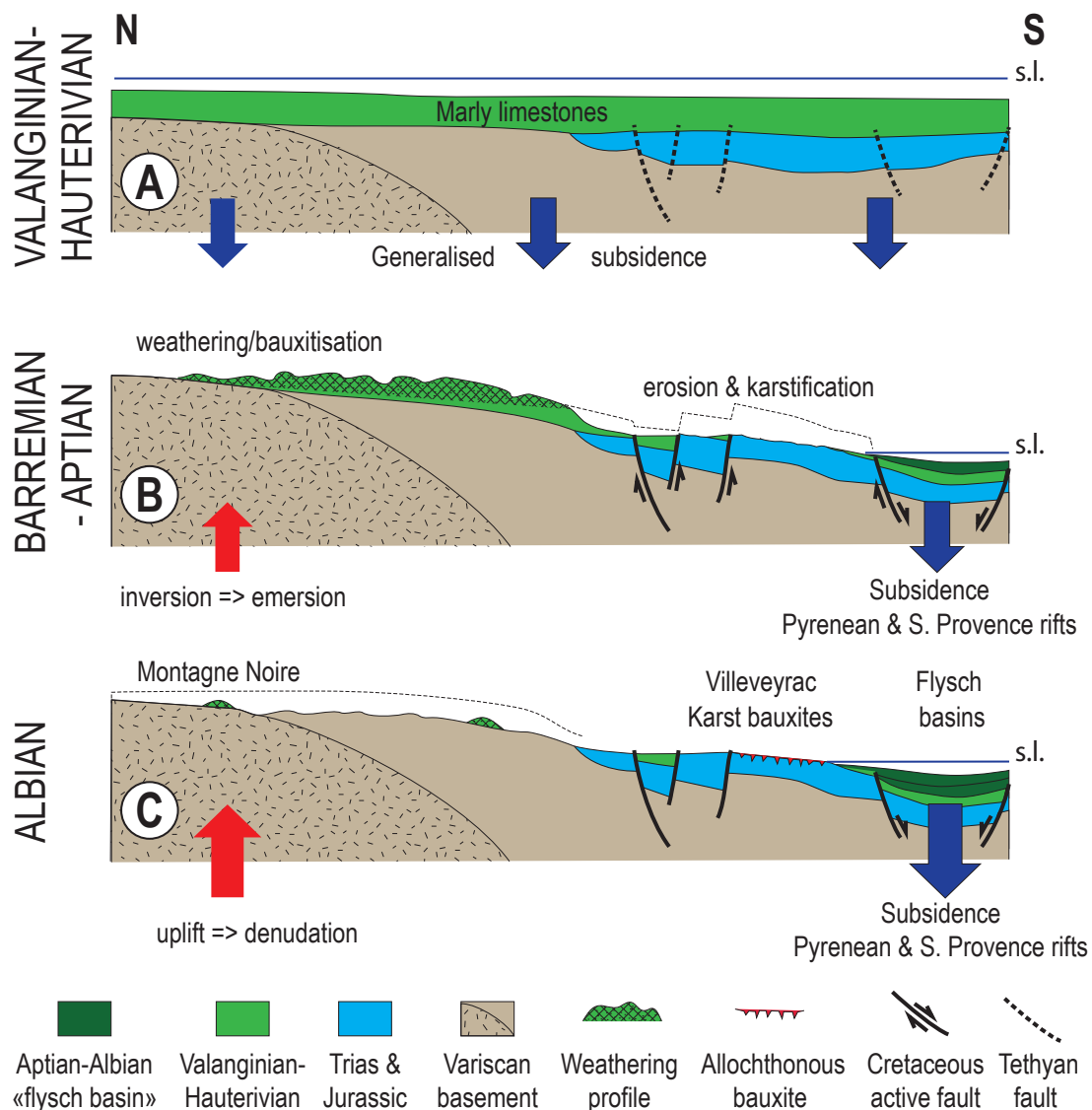


Fig. 13: Sequential conceptual north-south cross-sections illustrating the upper crustal structures and vertical movements, across of the study area (approximate location on Fig. 12), from Valanginian to Albian. Not to scale. Size of the subsidence/uplift vectors is related to their value. See text for explanations.

During Albian time (Fig. 13C), subsidence increased eastward (Fournier, F. *et al.*, 2016) and southward (Chelalou, R. *et al.*, 2016) of the study area, and uplift of the hinterland enhanced denudation (Barbarand, J. *et al.*, 2001; Peyaud, J. B. *et al.*, 2005; Olivetti, V. *et al.*, 2020). Warm and humid tropical conditions persisted and alteration was still going on (Chanvry, E. *et al.*, submitted) while the relative instability caused by the uplift allowed a topography gradient to evacuate weathering products, including reworked zircons. This situation lasted until the sedimentary cover was eroded, thus exposing the Variscan basement, which, consequently, provided an additional and increasing source for bauxite deposits. The uplift became more active and primary bauxites were then eroded and transported downstream towards karstified Jurassic traps.

8- Conclusion

The karst bauxite of Languedoc (southern France) was deposited during the mid-Cretaceous. These deposits are associated with a regional erosional hiatus, which obliterates most of the Cretaceous stratigraphic record of the area. LA-ICP-MS U-Pb analyses of 671 detrital zircons from 6 samples collected in three bauxite basins and their surroundings, established the following results:

- Detrital zircons of Late Aptian-Early Albian age, of probable volcanic origin, provide a maximum age for deposition of 115.5 ± 3.8 Ma. Together with the recent discovery of Late Albian foraminifera in the bauxite cover, we find that Languedoc bauxite deposits are Early to Middle Albian in age. This new results constrain the shortest stratigraphic interval for bauxite deposition in southeastern France
- Broadly similar age spectra were obtained for the 4 bauxite samples, covering an almost continuous range of ages from Late Variscan (circa 300 Ma) to Archean (3500 Ma). Bauxite age spectra include a Variscan peak at 350 Ma, a broad Early Paleozoic to late Neoproterozoic peak centred around 600 Ma, and significant clusters of late Mesoproterozoic (1 to 1.3 Ga) and late Paleoproterozoic (1.65 to 1.9 Ga) ages, and finally, a wide cluster of Neoproterozoic ages centred around 2.7 Ga. Such diversity suggests multiple reworking of detrital zircons through several cycles of erosion, transport and deposition. The large proportion of Late Mesoproterozoic to Archean ages found in the bauxite cannot be accounted for by a source from the Variscan basement (metasediments as well as crustal material), where these ages are poorly represented.
- Results suggest two distinct sources: i) reworking of a generalized sedimentary source, of regional extent, and ii) a source localised in the nearby Variscan basement high of the Montagne Noire, which was progressively uplifted during the Aptian and eroded during Albian.
- The range of ages found in the detrital zircons from the bauxite, which is wider and more complete than the sources in the Variscan basement, suggests multiple reworking of detrital zircons through several cycles of erosion, transport and deposition. It is believed that the sedimentary source (Valanginian marls) reworked older, post-variscan sedimentary sequences (e.g. Triassic continental sandstones) which contained detrital zircons, themselves reworked from older and more distant sources. The primary origin and routing of these old detrital zircons are still an outstanding question.

Interpretation of these new results in the light of previously published AFT data, suggests that the regional sedimentary source most probably corresponds to the thick sequence of early Neocomian (Valanginian) marls and marly limestones, which extended across southern France. Continental weathering of such sediments under warm and humid tropical climate promoted bauxites, which were then reworked, transported, and trapped in the karstified Malm. Dating of the very scarce detrital zircons found in the pristine Valanginian marls preserved in some down-faulted zones fits with the age spectra found in the bauxite samples; however, much larger samples of marls need be processed in order to establish statistically sound results.

Languedoc bauxites were deposited and preserved close to the base level, at the hinge zone between uplifted hinterland made of Variscan basement high and the subsiding basins in the south.

The new, tighter, age control on the deposition age of the allochthonous bauxites of Languedoc allows to correlate this event with early to mid-Cretaceous geodynamics affecting the southwest European plate. They correlate with i) the lithospheric rifting in the future Pyrenees and its eastward extension, offshore Provence, and ii) with the uplift and denudation of the southern French Massif Central and its SE-ward extension ("Durancian Isthmus"). Such deformations are probably different expressions of a left-lateral wrenching occurring along lithospheric plate boundaries, between SW Europe and Iberia with related continental blocs, to the south (Tavani, S. *et al.*, 2018).

The new age constraint (Early to Middle Albian) of the bauxites, allows new chronological constraints for this event. The hinterland uplift was first moderate during Barremian-Aptian, which allowed weathering and in-situ bauxitization. Uplift rate increased during Albian, which lead to dismantling and reworking of the bauxite. Finally, uplift had stopped by the end of Albian, which is recorded by the marine transgression over the bauxite deposits. This may mirror the evolution of the Pyrenean rift, with a diffuse and moderate rifting at the initial stage, followed by rift climax, reaching mantle denudation stage during the Albian (Lagabrielle, Y. *et al.*, 2010). Similarly, basin subsidence rate and flysch deposition in the Pyrenean rift seems to reach a maximum during the Albian (Souquet, P. *et al.*, 1985; Debross, E.-J., 1990) (Chelalou, R., 2015; Chelalou, R. *et al.*, 2016). In the less well resolved offshore mid-Cretaceous sequence off-shore Provence (Fournier, F. *et al.*, 2016), it could be hypothesized that the maximum accommodation was reached during the Albian.

This detrital zircons study from the Languedoc bauxites should be extended across the whole outcrops of bauxites in southeastern France, and compared with data from the mid-Cretaceous marine sediments.

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Supporting Information

Additional supporting information may be found in the online version of this article

Appendix 1 : Location of the analysed samples.

Appendix 2 : Analytical results: LA-ICP-MS U-Pb data for detrital zircons.

Appendix 3 : Concordia diagrams computed from LA-ICP-MS U-Pb data for detrital zircons