

The Paleogene continental basins from SE France: new geographic and climatic insights from an integrated approach

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Abstract

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- New insights into the geodynamic evolution, the paleogeographic framework and the
- 21 paleoclimatic setting of the Upper Eocene-Oligocene lacustrine basins from South-East France
- 22 have been provided by an integrative approach combining geochronology, micropaleontology

and stable isotope analyses. U-Pb dating on calcite, pollen-based climatostratigraphy and 23 strontium and sulfur isotope signatures of sulfates provided a robust chronostratigraphic 24 framework for the Upper Eocene and Oligocene succession from the Vistrenque, Alès and 25 Roussillon basins. The thick Priabonian- lower Rupelian lacustrine succession from the 26 27 Vistrengue Basin is interpreted to be deposited in a strike-slip basin associated with the latest stages of pyrenean shortening. Floral assemblages are used to reconstruct the vegetation cover 28 29 in the lacustrine systems and their neighbouring lowlands and highlands. Composition of saltwater inferred from strontium and sulfur isotope signatures of sulfates, the occurrence of 30 calcareous nannofossils, dinoflagellate cysts and pollen grains of mangrove-related plants 31 32 provided strong evidence for marine intrusions into the Alès Basin during the Priabonian and the Vistrenque Basin during the Rupelian and Chattian. Paleogeographic reconstructions of 33 Southeast France during the Priabonian and late Rupelian strongly suggest marine incursions 34 35 from the Alpine Sea via two potential pathways: upper Rhône Valley via the Crest Sill and lower Rhône Valley via the Haut-Var Pyrenean synclines. On the other hand, Upper Oligocene 36 evaporite basins in the Camargue area have been subjected to marine intrusions from the Western 37 Mediterranean Sea following the collapse of the eastern pyrenean relief during the Liguro-38 Provençal rifting stage. Finally, pollen analysis and Climate Amplitude Method revealed a brief 39 40 but significant stage of cooling and aridification at the very base of the Rupelian which could represent the regional and terrestrial expression of the global climate cooling following the 41 42 Eocene-Oligocene Transition.

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Keywords: Priabonian, Rupelian, Eocene-Oligocene Transition, U/Pb dating, pollens,
 strontium and sulfur isotopes, climatostratigraphy, marine intrusion.

1. Introduction

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48 In Southeast France, several continental basins were formed during the Paleogene and are 49 located in Languedoc (e.g. Alès, Saint-Chaptes and Issirac (ASCI) basins, Sanchis and Séranne, 50 2000; Lettéron et al., 2022; Séranne et al., 2021), Rhone Valley (Mormoiron Basin, Triat and Truc, 1974), Provence (e.g. Apt-Manosque-Forcalquier Basin, Lesueur, 1991; Marseille Basin, 52 Nury, 1990), and Camargue (e.g. Vistrengue Basin, Benedicto et al., 1996). These basins are 53 characterized by thick siliciclastic, carbonate and evaporite sedimentation (e.g. Rouchy, 1997). 54 The formation and the filling of these basins initiated during the Late Eocene (Priabonian) and lasted until the Late Oligocene (Chattian). These basins were interpreted as branches of the 56 European Cenozoic Rift System (ECRIS, Fig 1A) (Ziegler, 1992) or resulting exclusively from 57 the Oligocene to Aquitanian extension linked to the Liguria-Provence rifting (e.g., Vistrenque 58 Basin, Benedicto et al., 1996), and a few cases as polyphased extensional structures (e.g. 59 Sanchis and Séranne, 2000) (Fig. 1B, 1C). In their recent work, Séranne et al. (2021) reassessed 60 the origin of Priabonian continental basins located in Languedoc, between the Northeast trending Cévennes and Nîmes major regional faults and found that those basins resulted from left-lateral strike-slip movements of these faults which accommodated both the Western 62 European rift system extension (ECRIS) and the still active N-S shortening of the Central and 64 Western Pyrenees. The origin of evaporites in the saline paleolakes within the Paleogene basins of Southeast 66 France is poorly understood and is still a matter of controversy that has lasted for decades particularly on the origin of the salt water that may result from marine incursions and/or the 68 recycling of Triassic evaporites (Giraud, 1902; Fontes et al., 1996; Bodergat et al., 1999; Sissingh, 2001, 2006; Dèzes et al., 2004; Ziegler and Dèzes, 2007; Briot, 2008; Lettéron et al., 69 70 2017). Questions also remain on the patterns of saline water masses connections and diachronism of the evaporite intervals between the different basins but also on the role of changing climate on evaporite deposition and continental depositional system evolution at the key Eocene-Oligocene transition (e.g., Sissingh, 2001; Lettéron et al., 2022) which represents a crucial period for the Cenozoic global climate evolution. Until recently, assumptions have been made about the role of the Camargue area as a saline water corridor into the Languedoc

and Rhône Valley continental basins (e.g., Lettéron et al., 2018).

Since the Camargue area is located in a pivotal position among these France south-eastern basins, between the Languedoc basins to the west, the Provence basins to the east, and the Valence and Bresse grabens to the north (see location in Fig. 1A), the Paleogene Vistrenque Basin occupies a key geographic position for constraining the geodynamic and structural evolution of southeast France and Gulf of Lion areas during the Late Eocene and Oligocene times and provides new insights into these hitherto unresolved paleogeographic issues.

The Vistrenque Basin is considered as the deepest (~ 5 km) Cenozoic basin in southeast France and in the Gulf of Lion margin (Benedicto et al., 1996). The Paleogene sequence, up to 3200 m-thick, is entirely buried beneath the Rhône Delta sediments (Valette and Benedicto, 1995) (Fig 2). The drill cores have yielded very scarce and poorly preserved biota thus preventing the definition of a detailed and well-constrained chronostratigraphic framework. The correlation of thick halite and anhydrite deposits (Valette and Benedicto, 1995) with the Priabonian and Oligocene evaporites from the neighbouring Rhone and Languedoc basins is not yet established, thus preventing the reconstruction of a well-constrained paleogeographic map of these basins during the Paleogene and the elucidation of the question of their potential connection with the marine realm.

Comprehensive and detailed analyses of subsurface data from the Paleogene sedimentary record of the Vistrenque Basin (cores, cuttings, well logs) as well as from the neighbouring Alès Basin were undertaken to resolve these tectonic and paleogeographic issues (Fig. 3, Fig. 4). Based on the integration of different dating methods such as U-Pb absolute dating on

carbonates, pollen-based climatostratigraphy, the interpretation of strontium and sulfur isotopes measurements, and additional micropaleontological analyses (dinoflagellate cysts, calcareous nannofossils), the present work aims at: (1) constraining the lithostratigraphic and chronostratigraphic framework of the Paleogene sedimentary succession from the Vistrenque Basin; (2) bringing new insights into the pending issue of of the origin of salts in the Paleogene saline lakes of Southeast France and the occurrence of marine incursions, and (3) refining the paleogeographic and paleoclimatic reconstructions of Southeast France during the Late Eocene and Oligocene times. In order to complement the overview in time and space of the regional vegetation and climate evolution during the Oligocene, pollen data from the Canet 1 borehole from the Roussillon Basin (Eastern Pyrenees) (location in Fig. 1B) have been integrated in this study.

2. Geological setting

2.1 Regional tectonic setting and stratigraphy

Following the development of Upper Cretaceous carbonate platforms in Southeast France, the Mesozoic to Eocene cover has been folded and thrusted during the Pyrenean shortening which resulted from the convergence and collision of the Iberia-Corsica-Sardinia block with the European plate (e.g. Arthaud and Laurent, 1995; Séranne et al., 1995; Bestani et al., 2015). In Languedoc, the Nîmes and Cévennes faults have been shown to accommodate sinistral strikeslip movements as early as the Priabonian in response to both the Pyrenean shortening and Western European Rift extension (Séranne et al., 2021). As a consequence, within a 50 km wide wrenching zone located between the Cévennes and Nîmes faults, fluvial and lacustrine sedimentation occurred during Priabonian times, in releasing bends and oversteps of left-lateral strike-slip faults, (Séranne et al., 2021). The pyrenean orogen and associated syn- and tardi-

orogenic basins have been It was later dismantled from the late Rupelian to the Aquitanian to form the Gulf of Lion rifted margin (Arthaud et al., 1981; Arthaud and Séguret, 1981; Gorini et al., 1994; Séranne et al., 1995; Mauffret and Gorini, 1996; Mascle and Vially, 1999; Séranne, 1999; Guennoc et al., 2000; Lacombe and Jolivet, 2005). The Oligocene-Aquitanian Gulf of Lion rifting phase accompanied the retreat of slab and is followed by the Burdigalian break-up. Paleogeographic reconstructions made by Benedicto et al., 1996 suggest a high relief, more than 1 km of elevation, at the southern margin while the total extension across the Vistrenque Basin is estimated to range from 9 to 9.5 km. The break-up event was dated to the early to middle Burdigalian (Oudet et al., 2010a) and corresponds to the onset of the spreading of the Liguro-Provençal Basin which led to the south-eastward drift of the Corsica-Sardinia Block (Gorini et al., 1993; Séranne et al., 1995). During the Burdigalian, a marine transgression followed the break-up and Burdigalian to Tortonian post-rift marine sediments were deposited (Fig. 2B-C-D). By the end of the Miocene, the Messinian Salinity Crisis resulted in valley incision into the Miocene deposits and affected the Oligocene deposits in places. The resulting canyons are overlain by Pliocene and Quaternary sediments (Beaufort, 1954; Benedicto et al., 1996; Leroux et al., 2017).

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2.2 The Paleogene Vistrenque Basin

The Camargue region is constituted of two main Cenozoic basins separated by the Mesozoic Albaron High (Fig. 2A-C): the Vistrenque Basin to the West and the Vaccarès Basin (=Petit-Rhône Basin) to the East. The Vistrenque Basin is oriented NE-SW and extends over 50*30 Km²s This basin is linked to the northeast with the Pujaut Through (Fig. 2A) along the bounding NNE-SSW trending major Nîmes Fault that separates the basin from the Mesozoic hills of the Garrigues region (Fig. 2C; Semmani et al., 2022). The Vistrenque Basin is bounded to the south by the Highs of Grau-du-Roi and Saintes-Maries-de-la-Mer (Fig. 2A). The basin is structured

by a set of syn-rift faults mostly parallel to the Nîmes Fault (Fig. 1C, Fig. 2B-C) that determine tilted blocks within the basin but also by transfer faults that separate different sectors with specific structural styles (e.g., Fig. 2B; Benedicto et al., 1996 and their figures).

The maximum thickness of the Cenozoic sedimentary succession of the Vistrenque Basin is known from the Pierrefeu 1 well (4920 m) where it rests on a Lower Cretaceous marine carbonate basement (Fig. 2B). The Cenozoic infill of the Vistrenque Basin is subdivided into poorly defined pre-rift series, syn-rift series attributed to the Oligocene-Aquitanian extension, and post-rift series of Burdigalian to Pliocene and Quaternary age (Fig. 2B-C-D; Valette and Benedicto, 1995; Benedicto et al., 1996). Below the Aquitanian deposits, the Paleogene strata have been subdivided classically into three main lithological units of hundreds to thousands of metres thick: from top to bottom: *Série Calcaréo-salifère* Formation, *Série Rouge* Formation and *Série Grise* Formation (Fig. 2D). These subdivisions have been adopted by Cavelier et al. (1984) in their synthesis on the Paleogene of Southeast France and these authors suggested an Eocene age for the lowermost parts of the *Série Grise* Formation based on the observed similarities in the sedimentary trends with the neighbouring basins.

The compartmentalization of the Vistrenque Basin by transfer faults results in different stratigraphic successions between the northern Pierrefeu sector and the southern Gallician sector (Fig. 3A-C). The depositional setting and stratigraphic architecture of the Vistrenque Basin has been recently reassessed by Semmani (2022). The Paleogene lithological succession at the reference well (Gallician 9) from the Gallician sector is composed of five distinct formations (Fig. 3A). 1) The lowermost *Série Grise* Formation is several hundred metres thick (the base of the Paleogene is not reached in this sector) and is made of a monotonous succession of sandstones (calcarenites), conglomerates, marlstones and less frequent evaporites deposited in lacustrine settings. 2) The overlying *Série Calcaire* Formation is around 250 m thick and is made essentially of sandstones (calcarenites) and calcisilities (siltstones), and a few monomictic

conglomerates at the base. These lithologies become scarce upward while variegated calcareous claystones and vacuolar limestones become more frequent, and the unit is capped by a major subaerial exposure surface. 3) The *Série Mixte* Formation is 42 m thick and characterized by alternation of dominantly polymictic conglomerates, siliciclastic sandstones, variegated calcareous claystones and limestones. 4) The *Série Rouge* Formation is made of alternations of siliciclastic-dominated sandstones and marlstones with frequent conglomerates at the base. 5) The *Série Calcaréo-salifère* Formation is around 300 m thick in the Gallician sector and constituted mainly of limestones and papyraceous marlstones at the base. The upper part of the unit is made of two sequences made of alternations of sandstones, marlstones, limestones, anhydrite and halite.

In Pierrefeu 1 well (Fig. 3A), the nomenclature is slightly different and the *Série Calcaire* and *Série Mixte* formations have not been recognized probably because of lateral variations of facies. The top of the Série Grise from the Pierrefeu 1 well likely corresponds to the *Série Calcaire* defined in the Gallician sector. Compared to the Gallician sector, the considerable thickness of the evaporite-rich *Série Calcaréo-salifère* Formation (~900 m) from the Pierrefeu sector results from the abnormal overlay of evaporite intervals caused by halotectonic gravity slides on top of the NW-dipping roll-over of the Nimes Fault, linked to the Oligocene-Aquitanian extension (Valette and Benedicto, 1995).

2.3 The Paleogene Alès and Saint-Chaptes basins

Located between the Nîmes and Cévennes faults, the Alès and Saint-Chaptes basins are characterized by a thick Priabonian fluvial and lacustrine succession (up to 900m) deposited during the latest stages of pyrenean compression (Séranne et al., 2021). The Priabonian succession is unconformably overlain by Chattian continental deposits accumulated in extensional setting related to the Gulf of Lion rifting (Sanchis and Séranne,

2000). The Alès Basin is located along the Cévennes fault with a NE-SW direction (25 km long, 5 km large), while the Saint-Chaptes Basin occupied by a saline lake (ASCI lake system) whose mixed carbonate, siliciclastic and evaporitic sedimentation has been exhaustively studied by Letteron et al. (2018; 2022). These authors subdivided the Priabonian deposits in six unconformity-bounded parasequences from SPr1 to SPr6 defined as transgressive-regressive (T-R) cycles (Fig. 4). The chronostratigraphy of the Priabonian deposits is well-constrained and based on mammalian (Rémy, 1985, 1994; Rémy and Fournier, 2003; Rémy and Lesage, 2005) and charophyte biostratigraphy (Feist-Castel, 1971; Lettéron et al., 2017) coupled with magnetostratigraphic analyses (Lettéron et al., 2022).

2.4 The Roussillon Basin: Canet 1 borehole

The Canet 1 well, located in the Roussillon Basin (location: **Fig. 1B**, **Table S1**), cuts through a 1785 m thick sequence of Quaternary to Permo-Triassic deposits overlying the Hercynian basement (Gottis, 1958). The lithostratigraphic succession and chronstratigraphic framework of well Canet 1 are summarized in **Figure S1**. This section displays around 700 m of sandy-clayey-lignitic deposits (1015–1716 m depth) which may correlate the Late Oligocene to the Early Miocene interval (Gottis, 1958) and thus has been ascribed to the rifting episode of the Gulf of Lion evolution (Mauffret et al., 2001; Séranne, 1999; Romagny et al., 2020).

3. Material and methods

This work is based on the analysis of cores, well cuttings and well logs from boreholes drilled in the Gallician and Parrapon sectors of the Vistrenque Basin, boreholes and exposed sections from the ASCI system and cuttings from Canet 1 well located in the Roussillon Basin (see locations in Fig. 1B, Fig. 3 and Fig. 4; Supplementary data, Table S1).

3.1 Chronostratigraphic framework construction

The chronostratigraphic framework of the studied basins (Vistrenque, Alès, Saint-Chaptes and Roussillon basins) has been defined or improved by using U-Pb dating on carbonates, pollen-based climatostratigraphy and calcareous nannofossil biostratigraphy.

3.1.1 U-Pb geochronology

Sampling

The *Série Grise* Formation being almost entirely detrital, the U-Pb method has been used exclusively for calcite veins. In contrast, the overlying formations are carbonate-rich and display various carbonate mineral phases (cements, pedogenic carbonates, root calcifications, molluscan shells *-neomorphs*) making the dating method possible. The dating of these calcite phases, and therefore the corresponding geological events (e.g. subaerial exposure) helps to approximate the depositional age of the sedimentary intervals. Events post-dating the deposition give younger ages whereas events pre-dating the following depositional unit give maximum ages.

Analytical method

In order to carry out U-Pb dating on calcite, 30 samples containing a calcite mineral phase were selected for the confection of thin sections. For each sample, a 30 μ m-thick and a 100 μ m-thick thin section have been prepared respectively for petrographic observations and for the sampling by laser ablation for spectrometric analyses.

Prior to the analyses, sedimentological and diagenetic observations were carried thoroughly in order to identify the relevant mineral phases that should be dated and to avoid sampling from two different mineral phases, since sampling from different calcite generations would result in misinterpretations of ages. Diagenetic analysis and carbonate phase selection were performed by coupling cathodoluminescence and fluorescence microscopy. Confocal microscopy observations have been performed at the PRATIM (Aix-Marseille University) using an Axio-observer Z1 inverted confocal microscope with single or two-photon laser scanning comprising 04 visible lasers (Argon multi-ray laser: 458, 488 and 514 nm, DPSS 561 nm, He-Ne 594 nm, and He-Ne 633 nm) and a Chaméléon Vision II IR laser with pre-compensation system coupled with a ZEN 2010 software. Cathodoluminescence observations have been made at the CEREGE (Marseille, France) using a Cathodyne cold-cathodoluminescence from NewTec ® operating at 16 to 18 Kv and 160 to 200 μA coupled to an Olympus BH-2 microscope and Olympus-UPMTVC camera.

Once a carbonate phase has been individualized, the next step consists of the screening on this area for U and Pb content on thin section in order to estimate the variability of U/Pb ratios. For this purpose, elemental mapping of U, Pb, Fe and Sr was performed on the area of interest of the thin section. U-Pb dating on calcite used *in situ* Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) method which requires scattered U/Pb ratios in order to make robust statistical calculations thus allowing a reliable age, with the least uncertainty, to be obtained. Analyses were performed at the DatCarb A*midex platform (CEREGE, Aix-Marseille university).

Where the screening shows suitable U and Pb concentrations variability, about 30 circular spots of 100 µm diameter are picked within the mineral phase to perform the Laser Ablation. Calcite ablation on thick sections was performed using an ESI (Electro Scientific Industries) laser (193nm ArF Excimer laser) the resolution of which is 100 µm. Reference standards were used to correct the inter-elemental fractionation. The raw data were processed with the Lolite ® software designed for laser ablation analyses. An Excel macro, inhouse to the CEREGE, is used to handle the U-Pb data. Finally, the U-Pb age is calculated by using the Tera-Wasserburg diagram with add-on isoplot 4.15 (Excel). The obtained absolute ages have been calibrated on the Geological Time Scale of Gradstein *et al.* (2012). Further details on the methodology and the analytic parameters are given in Godeau *et al.* (2018) and their online supporting data section.

3.1.2 Pollen-based climatostratigraphy

Pollen grains have been extracted at the GeoBioStratData.Consulting laboratory (Rillieux-la-Pape, France) and the results have been integrated within a regional climatostratigraphic chart (e.g., Suc, 1984; Suc *et al.*, 2018, 2020). 52 samples of about 20-30 grams weight have been processed in order to obtain pollen floras using the classical method (acid digestions with hydrochloric acid (HCl) and hydrofluoric acid (HF), concentration in ZnCl₂ at density 2, and sieving at 10 µm). Only 18 samples yielded pollen grains in enough quantities (their geographic distribution is shown in **Figure 1B** and specified in **Supplementary data**, **Table S3**). The residues are mounted within glycerol between the slide and coverslip that allows to slightly move each pollen grain for observation purposes under various sides. Some pollen records, from the wells (Gallician 1, 4 and 9, Maruéjols 1) and the exposed Monteils section (Saint-Chaptes Basin, ASCI system), display a number of taxa that is lower than 25 (= systematic entities equivalent to families or genera). This value is the diversity threshold to assess the

representativeness of a pollen flora sample. These non-representative samples should be interpreted with caution.

The main advantage of this new pollen analysis approach based on a detailed taxonomic investigation is that it aims to reconstruct the botanical flora and is therefore a peculiar approach for the Paleogene times. The accurate examination of the morphological characters of fossil pollen grains and their comparison with modern pollen grains of rich and varied collections (ISEM mainly, GeoBioStratData.Consulting) and iconographic databases allow to confidently identify fossil pollen grains at least at the family level and very often at the genus level. A powerful ecological interpretation of pollen floras results from such botanical identifications. These botanical identifications are robust as it has been demonstrated that almost all the modern genera of plants already existed since the Early Paleogene and were altitudinally-latitudinally distributed similarly to the present time (Suan et al., 2017; Salpin et al., 2019; Suc et al., 2020). The significance of our pollen records in terms of ecological-climatological changes are also enhanced thanks to pollen countings. To date, pollen counting approach has been a routine investigation for the Pliocene and Quaternary and less used for ancient series.

High-quality in botanical pollen identification and pollen countings have allowed to successfully apply on these pollen floras the 'Climatic Amplitude Method' (Fauquette et al., 1998) in order to quantify the Paleogene paleoclimatic conditions (Suan et al., 2017; Salpin et al., 2019; Suc et al., 2020). The analyses have been performed on a AX10 Zeiss light microscope at X250 magnification, each pollen grain being observed at X1000 magnification for its identification.

A minimum of 150 pollen grains were counted per sample extra those of *Pinus* (a cosmopolitan genus), those of meso-microthermal trees, inhabiting mid-altitudes (*Cathaya*, *Podocarpus*, *Fagus*, *Cedrus*, *Tsuga*), and those of microthermal trees, inhabiting higher altitudes (*Abies* and *Picea*). The detailed pollen analyses are provided in **Supplementary data**

(Table S3). Taxa are grouped according to the climatic and/or ecological significance of their modern representatives (Table S3), resulting in two types of synthetic pollen diagrams. The first type includes all the pollen grains excepted those of *Pinus* and provides a local and regional image of the vegetation (including that growing on reliefs); the second type excludes the midand high-altitude plants and provides a perception of the local vegetation (i.e., that of low altitudes). In such diagrams, plants are grouped mainly according to the classification of Nix (1982) based on the mean annual temperature (MAT), the concerned taxa usually requiring relatively elevated to high precipitation: megatherms (MAT > 24 °C), mega-mesotherms (20 $^{\circ}$ C < MAT < 24 $^{\circ}$ C), mesotherms (14 $^{\circ}$ C < MAT < 20 $^{\circ}$ C), meso-microtherms (12 $^{\circ}$ C < MAT <14 °C), and microtherms (MAT < 12 °C). The other groups have various significations: environmental for the herbaceous plants (more often not-identifiable at the infra-family level), ecological for the water plants, indicative of some dryness (Mediterranean sclerophyllous plants, steppe plants and subdesertic plants), insignificant (for example, the Cupressaceae with a Cupressus-Juniperus like pollen unidentifiable at the genus level). Details about taxa composing the groups are provided in Table S3. Reworked palynomorphs from older periods are not very abundant except in few samples (Canet 1 1100 and 1701 m; Gallician 1 1648 m; Gallician 9 2530 m; Table S3). They are easily recognizable by their morphology and/or their darker colour and/or their altered preservation. The varying content of successive pollen locations is mostly considered as markedly driven by global climate changes which was affected by major and secondary fluctuations during the time interval 38–23 Ma as illustrated by the global oxygen reference isotope curve (Westerhold et al., 2020). Accordingly, pollen floras are hypothesized to correlate with the global climatic evolution, which constitutes the principle of climatostratigraphy (Suc, 1984; Suc et al., 1995, 2018, 2020; Popescu et al., 2010). Factually, we propose to correlate each pollen assemblage

with the oxygen isotope stage that corresponds the best to its climatic significance, still within

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the established stratigraphic succession and/or within the chronologic range separately indicated by mammal fauna or calcareous nannofossils when available.

This approach contrasts with the traditional palynostratigraphy based on the direct attribution of a chronological value to some pollen grains (Sittler *et al.*, 1975; Châteauneuf, 1980; Châteauneuf et al., 2019) that may lead to serious misinterpretations (see the **Supplementary Data**).

3.1.3 Calcareous nannofossils

The analysis of calcareous nannofossils has been performed for refining the chronostratigraphic framework and evidencing potential marine incursions. Calcareous nannofossils have been studied in the fraction of 2–30 µm and separated by decantation method using a 7% solution of H₂O₂. Smear-slides were mounted with Canada balsam and analyzed with an Olympus transmitting light microscope at x 1200 magnification. Nannofossil identifications follows the taxonomic classification of Perch-Nielsen (1985) and Raffi et al. (2006). Nannofossil zones are after Martini (1971). Further calibration follows Raffi et al. (2006) and Gradstein et al. (2012).

3.2 Climatic Amplitude Method

The endeavour in pollen identification and counting implies the possibility to use the Climatic Amplitude Method to reconstruct climate conditions during the deposition of the sediments. This method allows estimating past climate variables by transposing the climatic requirements of the maximum of modern taxa to the fossil records. This approach relies on the relationship between the relative pollen abundance of each individual taxa and climate, defined after comparison with more than 6,000 present-day pollen records distributed worldwide. This

method accounts not only for the presence/absence criterion but also pollen percentages to provide more reliable reconstructions (Fauquette et al., 1998). The estimated mean annual temperatures (MAT), mean temperatures of the coldest (MTC) and warmest (MTW) months and mean annual precipitation (MAP) concern low-elevation vegetation because mesomicrothermal and microthermal taxa (Cedrus, Fagus, Tsuga, Cathaya, Podocarpus Abies and *Picea* which live today at higher elevations in tropical-subtropical regions) were excluded from the process (but not from the percentages) to avoid a cold bias linked to transport from higher elevations. Pinus, which may inhabit different vegetation belts, is also excluded from the calculation. The excluded taxa were defined based on the distribution of vegetation today. Indeed, the types of vegetation deduced from our pollen data are found in South-eastern China today at around 25°N and 110 to 120°E (Hou, 1983). In this region, the vertical distribution of the vegetation is characterized, from the base to the top of the massifs by evergreen broadleaved forest, mixed evergreen and deciduous (Betula, Acer) broad-leaved forest, Cathaya/Tsuga forest, Picea/Abies forest and high mountain meadows (Hou, 1983). In the Climatic Amplitude Method, the estimates for each climatic parameter are given as an interval (minimum and maximum values of the parameter). A most likely value (MLV) is calculated, which corresponds to a weighted mean. The MLV has been statistically tested on modern pollen data and has provided reliable results (Fauquette et al., 1998).

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3.3 Geochemical modelling of strontium and sulfur isotopes on sulfates

Strontium and sulfur isotope compositions of gypsum and anhydrite have been proved to be reliable markers of the origin of the waters from which they precipitated and geochemical modelling approaches make it possible to quantify the proportions of different salt waters within a mixture (Denison et al., 1998).

3.3.1 Sampling and measurements

Five anhydrite samples from Pierrefeu-1 well (Vistrenque Basin, see position in Fig. 5) and three anhydrite samples from Les Fumades wells (SC-1 and SC-2, Alès Basin, see position in Fig. 4) have been selected for δ^{34} S and 87 Sr/ 86 Sr laboratory measurements.

Strontium isotope measurements were carried by the SARM-CRPG laboratory (Vandoeuvre-les-Nancy, France). Samples have been powdered ($<50 \,\mu\text{m}$), weighted and mixed with HNO₃ and HF and after evaporation, mixed with concentrated HCl. After complete digestion of the sample and Sr separation, $^{87}\text{Sr}/^{86}\text{Sr}$ was measured by using a TIMS Triton Plus (Thermo electron) in static mode. The standard NBS987 has been used as a reference. All measurements have 2σ error are lower than 0.000012.

S isotopic composition of sulfates from anhydrite and gypsum samples and elemental (native) sulfur was measured by combustion using an Elementar vario PYRO cube elemental analyzer (Elementar GmbH) in-line with an Isoprime 100 mass spectrometer (Manchester, UK) in continuous flow mode at the University of Burgundy in Dijon, France. Isotope data are expressed in delta notation and reported in units per mil (‰). The δ^{34} S data are reported with respect to the international standard Vienna Cañon Diablo Troilite (VCDT). Analytical errors are \pm 0.3 ‰ (2 σ) based on replicates for standard materials.

3.3.2 Modelling of strontium and sulfur isotope composition of mixed waters

The Sr isotope composition of two-component mixtures/endmembers has been described in detail by Faure (1986) and applied to Sr and S isotopes (Andersson et al., 1992; Denison et al., 1998). The isotopic ratio (87Sr/86Sr and 34S/32S) Im is computed as follows:

$$Im=[f1.C_1.I_1+(1-f_1).C_2.I_2]/[f_1.C_1+(1-f_1).C_2](1)$$

- Where I₁ and I₂ represent the isotopic ratios of the first and second end member, respectively.
- 411 C_1 and C_1 are the Sr or SO_4^{2-} concentration in the first and second end members, respectively
- and f1 is the volume fraction of the first end member water in the mixture.
- Various endmembers have been selected for the modelling: 1) seawater, 2) continental water
- 414 whose Sr and S content is controlled by the leaching of Triassic evaporites, 3) continental water
- whose composition derived from the leaching of metamorphic rocks.
- The Sr concentration of Paleogene seawater (15 ppm) is derived from the estimations by
- Vollstaedt et al. (2014); such a value is close to modern concentrations (~8 ppm). Sulfur
- 418 concentration was assumed to be similar to that of modern seawater (2800 ppm). The ⁸⁷Sr/⁸⁶Sr
- values for Priabonian to Chattian seawater range from 0.7077 to 0.7083 (Mc Arthur et al., 2012)
- 420 while δ^{34} S is assumed to be close to modern values (+20%).
- Riverine waters influenced by the leaching of Triassic evaporites have been considered to
- have retained the 87 Sr/ 86 Sr and δ^{34} S of the original gypsum or anhydrite. 87 Sr/ 86 Sr values ranging
- from 0.7076 to 0.7080 have been reported for Triassic (Keuper) evaporites from the French
- 424 Pyrenees (Albarède and Michard, 1987), Italy (Dinelli et al., 1999), Spain (Alonso-Azcarate et
- 425 al. 2006; Ortí et al., 2014), while δ^{34} S values range from +10 % to +17 % (Utrilla et al., 1992;
- 426 Dinelli et al., 1999; Boschetti et al., 2011).
- Riverine waters draining metamorphic or igneous bedrocks should have higher ⁸⁷Sr/⁸⁶Sr
- ratios as shown in present time in the Cevennes Massif, with values ranging from 0.712 to 0.714
- for waters draining granites and from 0.714 to 0.717 for waters draining schists (Petelet-Giraud
- et al., 2006). Regarding sulfur isotope composition, magmatic and metamorphic rocks are
- known to be characterized by δ^{34} S ranging from -12 % to +12% (Chaussidon et al., 1989;
- 432 Giacometti et al., 2014).

4. Results and interpretation

4.1 U-Pb dating

Among the examined 12 samples, only three samples meet the conditions of the U-Pb dating method: (a) G9 13D, (b) G9 7, and (c) G9 K35. Relevant samples and associated ages are summarized in Table 1.

4.1.1 G9 13D Sample: subaerial exposure on top of the Série Calcaire

In this sample, microstalactitic calcite (A1) lines the walls of a dissolution vug. It is post-dated by an anisopachous calcite cement (A2) which is in turn post-dated by an isopachous equant sparry calcite cement (I1) (Fig. 6A). Only cement A1 displays dull luminescence while the following cements are non-luminescent and thus cannot be distinguished under cathodoluminescence. However, observations under confocal microscope made it possible to distinguish the cement generations (Fig. 6C). Uranium concentrations are very low in the A2 and I1 cements thus precluded dating analyses (Fig. 6B). However, the cement stage A1 displays enough U and Pb thus allowing the dating method to be applied (Fig. 6D). The calculated age of the early diagenetic microstalactitic cement A1 is 31.27 +/- 1.90 Ma which gives a early to middle Rupelian age after GTS2012 (Gradstein et al., 2012).

4.1.2 G9 7 sample: vadose cement within the lowermost interval of the Série Rouge

This sample displays anisopachous pendant microsparite cements, up to 600 μ m thick around clasts in a polymictic conglomerate (Fig. 7A). In this sample, carbone and oxygen isotope analyses on the calcite yielded very negative values ($\delta^{18}O = -5.11$ % V-PDB & $\delta^{13}C =$

-5.59 ‰ V-PDB) thus indicating meteoric origin of the precipitation water. In addition, the anisopachous nature of the cement suggests precipitation under vadose meteoric conditions. Such cements represent the earliest phase of cementation and formed prior to burial and mechanical compaction. The cement therefore likely formed shortly after deposition and infill of the fluvial channel, in a vadose environment within an alluvial plain. U and Pb concentrations are favourable thus making it possible to test the dating method. 74 spots have been used and the concentrations show sufficient dispersal of the U/Pb ratio thus enhancing the accuracy of the method (Fig. 7B). The U-Pb age found for this carbonate phase using the Tera-Wasserburg diagram is 27.88 +/- 1.71 Ma which indicates a late Rupelian to early Chattian age according to the GTS (Gradstein et al., 2012). The initial composition ratio of ²⁰⁷Pb/²⁰⁶Pb is 0.8321 +/- 0.034.

4.1.3 G9 K35 sample: calcite veins within the lower Série Grise

The K35 sample is from the terrigenous-rich *Série Grise* Formation. In this sample, the calcite analysed for U-Pb dating consists of the filling of fractures (veins) within the silty marlstones host rock (**Fig. 7C**). The vein is about 1 mm wide and it is filled with equant to fibrous calcite, the crystals sizes average 200 µm.

Under cathodoluminescence, the calcite vein is non-luminescent (**Fig. 7D**). In the calcite vein, U and Pb concentrations are favourable to the dating method (**Fig. 7E**). The calculated Tera-Wasserburg U-Pb age for the calcite vein infill is 21.46 +/- 2.19 Ma.

4.2 Calcareous nannofossils

Calcareous nannofossils are recorded in seven samples (Supplementary data: Table S2), supporting some marine water incursions into the Alès Basin during the Priabonian (crossroad

D716/D981 sample), in agreement with the occurrence of dinoflagellate cysts and mangrove pollen grains. Some other marine water incursions are indicated in the Gallician 1 borehole at depths 1810 m (from the lowermost *Série Calcaréo-salifère* Formation ascribed to late Chattian) and 1594 m (ascribed to earliest Aquitanian), consistently with the occurrence of marine dinoflagellate cysts in the studied samples from the Canet 1 borehole. The calcareous nannofossil assemblages identified from the Alès and Vistrenque basins (Crossroad D716/D981, Maruéjols 1 433 m, Gallician 1 1810 m and 1594 m) are poor in species with a short biostratigraphic range: only *Reticulofenestra bisecta* and *R. lockeri* point out a chronological interval from ca. 38 Ma to ca. 23 Ma (Anthonissen and Ogg, 2012 in Gradstein et al., 2012) i.e., covering the time interval investigated in this study including the Priabonian and the entire Oligocene. More accuracy is provided by the calcareous nannofossil assemblages from the Canet 1 borehole (Table S2): sample at 1701 m can be referred to biozones NP23–NP24 (32–26.8 Ma), sample at 1470 m to biozones NP24–NP25 (29.7–23.2 Ma), sample at 1100 m to biozones NP25–NN1 (26.8–22.8 Ma) (Martini, 1971; Raffi et al., 2006; Anthonissen and Ogg, 2012).

4.3 Pollen-based reconstructed paleoclimates and climatostratigraphy

4.3.1 Results

A total of 122 taxa have been identified in the 18 pollen records (**Table S3**). These pollen floras show prevalent mega-mesothermal plants and, secondarily, mesothermal plants and the conifers *Cathaya* and *Podocarpus*. Megathermal elements are mostly present in the Priabonian samples where they show relevant variations. Steppe elements mainly occur in the Rupelian samples. The other plant groups are intermittently represented.

Pollen record of five samples from wells in the Vistrenque Basin (Gallician 1 1648m, Gallician 1 1775m, Gallician 4 1580m, Gallician 9 2530m, and Vauvert 1 3200m; Fig. 8, see location in Fig. 5) was relatively poorly diversified and poorly preserved. Indeed, these pollen grains have been partially vitrified and therefore some of their morphological characters (ornamentation, structure) are severely altered. A certain amount of pollen can still be identified even though a great percentage of undeterminable pollen grains may lead to lessen the significance of some of these microflora (percentage of unidentifiable pollen grains: Gallician 1 1648 m: 8.19%; Gallician 4 1580 m: 37.36%; Vauvert 1 3200 m: 10.81%).

In addition, some of these pollen floras are characterized by a low number of taxa (Gallician 1: 24 taxa at 1648 m, 19 taxa at 1775 m; Gallician 4 1580 m: 13 taxa; Gallician 9 2530 m: 20 taxa). Among the possible hypotheses to explain such biases in preservation and plant diversity (oxidation, transport, diagenesis), the usual occurrence of oil traces during the sample processing points to a thermal diagenesis in relation with sediment maturation within the oil window. In the Gallician samples, the dominant pollen grains are those of Gymnosperms which remain often identifiable despite their poor preservation, especially of meso-microthermal (*Cathaya*, *Podocarpus*, and some *Tsuga*) and microthermal (*Abies*) trees which may reflect the relief of the Pyrenees and subsidiarily of the Alps.

The pollen record of the Gallician 9 2530m sample shows the dominance of megamesothermal (Cupressaceae papillate pollen representative of the former 'Taxodiaceae' family) and both *Cathaya* and *Podocarpus* but a significant feature of this sample is the occurrence of some mesothermal plants, Cupressaceae (*Cupressaceae-Juniperus*-type) and *Ephedra* (steppe plants).

In **Figure 8**, the vertical arrangement of samples first follows the stratigraphic knowledge of the studied basins (Gottis, 1958; Clauzon, 1990; Séranne, 1999; Lettéron et al., 2017, 2018, 2022) and chronology is specified thanks to informations provided by (1) mammal faunas

(Depéret, 1917; Rémy, 1985; Rémy, 1994; Rémy and Fournier, 2003; Rémy and Lesage, 2005) and (2) calcareous nannofossils that have been searched in the samples containing frequent to abundant marine dinoflagellate cysts: they are specified below and in the **Supplementary data** (**Table S2**).

The climate has been quantified for all the pollen spectra except for Gallician1 1775 m, Gallician 4 1580 m and Saint-Jean de Maruéjols – Pit hill because of the low taxa diversity. The results are given, for each climatic parameter, as an interval and a Most Likely Value for each site (Table 2).

4.3.2 Interpretation

The analysed pollen records allow to reconstruct the local and regional vegetation, from low altitude to elevated reliefs (Fig. 8). Lower places were sporadically occupied by *Avicennia* mangrove with probably Rhizophoraceae, suggesting nearby marine environments, which are also supported by the record of marine dinoflagellate cysts. Occurrence of lakes is revealed by frequent freshwater plants (*Aglaoreidia*, *Myriophyllum*, Oenotheraceae, Restionaceae, etc.) and the *Botryococcus* microalga. The coastal environments and lake banks were also inhabited by riparian swamps with *Taxodium-Glyptostrobus* (i.e., illustrated by the *Taxodium*-type pollen grains). Lowlands were occupied by an evergreen subtropical forest rich in mega-mesothermal plants (*Alangium*, *Engelhardia*, *Embolanthera*, *Distylium*, *Nyssa*, *Mussaenda*-type, *Rhoiptelea*, Sapotaceae, *Symplocos*, *Uncaria*-type, Cupressaceae former 'Taxodiaceae', etc.) including some megathermal plants (*Amanoa*, *Bombax*-type, *Afzelia-Peltophorum*-type, *Canthium*-type, *Croton-Jatropha*, *Peltophorum*-type, *Mappianthus*, *Phyllanthus*, *Peltogyne*-type, *Sapindaceae*, *Thumbergia*, etc.). Vicinity of higher lands is attested by abundant mesothermal plants (*Acer*, Araliaceae, *Carpinus*, *Carya*, *Eucommia*, *Liquidambar*, *Parrotia*, *Pterocarya*, *Quercus*, *Ulmus*, *Zelkova*, etc.), which constituted a mesophilous deciduous forest. The development of

altitudinal forest vegetation (*Cathaya*, *Podocarpus*, *Cedrus*, *Tsuga*, *Abies*, *Picea*) suggests the occurrence of reliefs, especially of high reliefs to the south as evidenced by Canet 1 well and Gallician wells pollen floras.

Cupressaceae trees (*Cupressus-Juniperus*-type) may contribute to the lowland and higher vegetation as they can be represented by diverse taxa growing today in subtropical to high altitude forests, because of our inability to distinguish their pollen. Sclerophyllous Mediterranean elements (*Olea* mainly, *Phillyrea*, *Quercus ilex*-type) episodically developed, simultaneously with decrease in taxa requiring great humidity and increase in herbaceous taxa. It is possible to conceive that these plants already grew on drier soils and tolerated less humid periods. Drier conditions are revealed by development of steppe assemblages (mainly constituted of *Ephedra*), in parallel with reducing of taxa requiring humid climate.

Variability observed in the Priabonian pollen assemblages appears to be referred to climate fluctuations during a warmer phase. All these pollen records come from exposed sections and are not concerned by biases in preservation which often affect deep sediments from boreholes. They mainly consist in opposed amounts of thermophilous forest elements (megatherms and mega-mesotherms) and herbaceous and steppe elements (*Ephedra*) pointing to more open and drier environments. Variability observed in the Oligocene pollen assemblages could refer to climate fluctuations during a cooler phase resulting in alternations of drier and more humid episodes.

All the Oligocene pollen records come from drilled sediments in which pollen preservation may be poor, that makes uncertain their paleoenvironmental interpretation. However, these major (Priabonian–Rupelian transition) and minor (intra-Priabonian and intra-Rupelian–Chattian) changes displayed by pollen assemblages are consistent with the reference oxygen isotope curve (Fig. 8; Westerhold et al., 2020). For example, the sample Gallician 9 (2530 m) is understood as illustrating the major change in climate marking the Eocene–Oligocene

transition because of scarcity in megathermal plants in opposition to the abundance of *Ephedra*. Finally, the two samples Gallician 1 (1775 and 1648 m) are correlated with two successive warmer episodes during the late Chattian because of high amount of mega-mesothermal plants and the persistence of *Ephedra* in low percentage.

Paleogeographic reconstructions indicate the existence of elevated topography southwards to the Camargue and Languedoc basins (Benedicto et al., 1996). These reliefs supplied detrital sediments and pollen grains of high-altitude plants that have undergone low to moderate transport to the depositional areas of the Vistrenque and of the Alès basins. The abundance of *Podocarpus* in the Gallician samples, a marker of the Pyrenean flora during the Paleogene, indicates that the pollen of these samples is sourced from these highlands.

In the Alès Basin, the climatic deterioration at the Eocene-Oligocene transition is expected to follow the Montchamp pollen flora, indicating persistent warm conditions. The reconstructed MAT and MTC are very high for this location (Table 2). Because of their low percentage in megathermal plants in contrast to the occurrence of *Ephedra*, the pollen records of Monteils and of the Pit hill of Saint-Jean de Maruéjols attest less warm and weakly drier conditions than the Crossroads D716/D981, Célas, and Montchamp microfloras. The estimated MAT at Monteils is the lowest for this time period (Most Likely value around 18.7°C instead of 20 to 21°C, Table 2). Some cooling is also evidenced by the Serre de Cauvel pollen assemblage with respect to decreased megathermal plants and increased Mediterranean sclerophyllous elements. However, this cooling should be less important as it is not observed in the estimated climate. The Monchamp microflora contains *Avicennia* and Rhizophoraceae pollen grains. The latter are also recorded at Serre de Cauvel while *Avicennia* is also recorded at Saint-Jean de Maruéjols Pit hill, Célas Tunnel and Crossroads D716/D981. Mean temperature of the warmest month (MTW) is stable all along the period (Table 2). Euzet Hill, Serre de Cauvel, Crossroads D716/D981, Maruéjols 1 798 m, and Saint-Jean de Maruéjols Pit hill samples contain high

proportions of conifers (*Podocarpus*, *Cathaya*) that do not result from preservation bias because of their high floristic diversity (>30 taxa) but from high pollen supply from mid-altitude.

In the Vauvert 1 3200 m sample, the presence of relatively frequent pollen of Mediterranean sclerophyllous plants may point to the climatic cooling at earliest Oligocene, supported by few megathermal plants, reduced mega-mesothermal plants and increasing herbaceous open environments in a context of shortening freshwater bodies. This cooling and lower humidity are also well marked in the estimated mean annual temperature (MAT), mean temperature of the coldest month (MTC) and mean annual precipitation (MAP) (Table 2). The estimated MAT at Vauvert is in agreement with the value obtained at Saint-Lions, located 150 km northeastward of the Camargue, presently in the Digne thrust, for the same period (Fauquette et al., 2015). In the Gallician 9 2530 m sample, some aridification of the climate is suggested by the local expansion of *Ephedra* that leads to a low estimate of mean annual precipitation (Table 2). Pollen assemblages of Gallician 4 1580 m and Gallician 1 1648 m are consistent with a Late Oligocene age.

Our ascription of the studied interval of Canet 1 well (1701–1100 m) to Oligocene is based on the occurrence of above-mentioned calcareous nannofossils (**Table S2**) and corresponds to the presence-abundance of *Podocarpus* pollen, never recorded in Southern France since the earliest Aquitanian (Bessedik, 1984). Considering the calcareous nannofossil range, the sample Canet 1 (1701 m) is ascribed to a warmer and still dry episode during the late Rupelian because of increased percentage in megathermal plants and a significant occurrence of *Ephedra*. Similarly, the sample Canet 1 (1470 m) is referred to a cooler episode of the early Chattian as supported by the absence of megathermal plants and a relative high presence of *Ephedra* associated to *Calligonum*. The sample Canet 1 (1186 m) is located in an intermediate cooler episode of the mid-Chattian because of the absence of megathermal plants, a relatively weak occurrence of mega-mesothermal plants and the persistence of *Ephedra* in a low proportion.

Finally, the uppermost Canet 1 (1100 m) sample is ascribed to a lesser warm episode just before the Chattian–Aquitanian transition because of its lower amount of mega-mesothermal plants and greater percentage of mesothermal plants. Climate reconstruction of Canet 1 well show that MAT are 1 to 2°C and MTC are 1 to 5°C lower than during the Priabonian. MAP are also slightly lower than during the Priabonian (Table 2).

4.4 Geochemical modelling of the strontium and sulfur isotopes of sulfates

4.4.1 Pierrefeu-1 well (Vistrenque Basin)

Anhydrite samples from Pierrefeu 1 well display 87 Sr/ 86 Sr values ranging from 0.70777 to 0.708201 and δ^{34} S values comprised between +10.92 ‰ and +17.10 ‰ (**Table 3**). Anhydrites from core K2 fall within the domain of Triassic evaporites while samples from cores K5 and K6 display significantly higher 87 Sr/ 86 Sr values (**Fig. 9A**).

Mixing fluid models revealed that Sr and S isotope composition of anhydrites from Pierrefeu-1 is consistent with a formation from a mixed water composed of a riverine end-member whose Sr and S content is controlled by the leaching of Triassic evaporites and a marine end-member. The Sr and S concentrations and isotope composition of the volume fraction of each end-member for the best-case scenario, are given in Supplementary Materials (**Table S4**). Matching between mixed-water models and isotope measurements on anhydrites integrates the effect of fractional precipitation which may lead to a difference of up to +2 % in δ^{34} S between sulfates and precipitating water (Raab and Spiro, 1991).

4.4.2 SC-1 and SC-2 wells (Alès Basin)

Anhydrite from the Alès Basin (SC-1 and SC-2 wells) display a narrow range of 87 Sr/ 86 Sr values (0.70768-0.70790) while δ^{34} S is comprised between +12.11 ‰ and +26.98 ‰ (**Table 3**). The sample from SC-1 well fall within the domain of Triassic evaporites while samples from SC-2 exhibit δ^{34} S values which are significantly higher than those of Priabonian sea-water (**Figure 9B**).

The high $\delta^{34}S$ values of gypsum minerals from SC-2 most likely arise from isotope fractionation during microbially mediated reduction of sulfates. If sulfate reduction takes place in a closed basin, the $\delta^{34}S$ of residual dissolved sulfates will be higher compared to the original fluid (Utrilla et al., 1992; Palmer et al., 2004). Additionally, measurements on native sulfur formed within anhydrite-bearing horizons in SC-2 well yielded negative $\delta^{34}S$ values ranging from -5.99 ‰ to -6.43 ‰. These negative $\delta^{34}S$ values strongly suggest that the elemental sulfur from SPr3 evaporitic interval is a byproduct of sulfate-reducing bacterial activity (Lindtke et al., 2011).

As a consequence, although the SC-1 sample unequivocally indicates precipitation from a water influenced by the dissolution of Triassic sulfates, the bacterial sulfate-reduction processes affecting the anhydrites from SC-2 prevent to draw any conclusion regarding a possible mixing with marine water.

5. Discussion

5.1 Chronostratigraphic framework of the Paleogene of the Vistrenque Basin

Sedimentological and diagenetic studies, and petrographical observations on core from the Paleogene succession in the Gallician 9 well revealed than the top of the *Série Calcaire* and the

whole overlying *Série Mixte* are affected by repeated subaerial exposition (Semmani, 2022) as suggested by the occurrence of pedogenic limestones and karst features. The development of dissolution vugs and the subsequent formation of anisopachous calcite cements (meteoric vadose environment) in the sample G9 13D (Gallician 9: 1960 m depth) are linked to these repeated stages of subaerial exposition affecting the top of the *Série Calcaire* Formation and the whole overlying *Série Mixte* Formation. The 31.2 Ma age obtained for the subaerial exposures towards the top of the *Série Calcaire* (G9 13D) represents the oldest possible age for the deposition of the overlying *Série Mixte* Formation (Fig. 10).

The 27.88 Ma age obtained within an early vadose cement from a fluvial conglomerate, in Gallician 9 well at 1984 m, suggests a late Rupelian to early Chattian age for the lower part of the *Série Rouge* Formation. The difference between the two absolute ages (~ 3.4 Ma) corresponds therefore to the time deposition of the *Série Mixte* and lower part of *Série Rouge*. Such a long duration, compared to the preserved thickness (~ 45 m) is consistent with low sedimentary rates and the occurrence of hiatuses punctuating the sedimentation as suggested by subaerial exposures affecting the interval (Semmani, 2022).

In addition, pollen-based climatostratigraphic approach suggests that the sample from Vauvert 1 at 3200m deposited very close to the Priabonian to Rupelian transition, thus indicating likely a Late Eocene (Priabonian) age for the lowermost part of the *Série Grise* Formation (Fig. 10). In Gallician-9, the 31.2 Ma age obtained at 1960 m and the early Rupelian age at 2530 m suggest sedimentation rates greater than 200 m/Myr in the upper *Série Grise* and *Série Calcaire* interval. Correlations with Vauvert-1 (Fig. 5) suggest sedimentation rates ranging from 300 to 500 m/Myr. The base of the *Série Grise* being not reached at 3630 m in Vauvert-1, extrapolating such sedimentation rates below 3200 m would suggest that the formation covers large parts of the Priabonian – if not the entire stage.

The late Priabonian oolithic lacustrine carbonates evidenced north of the Vistrenque Basin (Semmani *et al.*, 2022) would therefore represent a marginal lake, lateral equivalent of the *Série Grise* Formation from the basin depocenter. The *Série Grise* Formation may also have deposited coevally with part or all of the Priabonian fluvial and lacustrine succession from the neighbouring Alès, Saint-Chaptes and Issirac basins (Lettéron *et al.*, 2022).

Pollen records from Gallician 4 1580m and Gallician 1 1648m suggest a Late Oligocene (early Chattian) age for the lowermost part of the *Série Calcaréo-salifère* Formation. These findings agree with Chattian to Aquitanian ages derived from the Sr and S isotope signatures of sulfates from this interval in Pierrefeu 1 well (Fig. 9). Based on these age constraints, the *Série Rouge* Formation is late Rupelian to early Chattian in age (Fig. 10). Finally, the Aquitanian absolute age (21.46 +/- 2.19 Ma) of the calcite vein affecting the *Série Grise* in sample G9-K35 from Gallician 9 well is consistent with the infill of a fracture formed during the Oligocene-Aquitanian rifting of the Gulf of Lion margin (Séranne et al., 1995).

5.2 Tectonic and geodynamic implications

The here-defined chronostratigraphic framework of the Vistrenque Basin evidences the occurrence of a thick Priabonian to lower Rupelian sedimentary succession, thus being coeval with the fluvial-lacustrine sedimentation occurring west of the Nîmes fault in the Alès, Saint-Chaptes and North Montpellier basins (Séranne et al., 2021; Lettéron et al., 2022). The regional mapping of syn-depositional structures within these basins evidenced a left-lateral shearing of the area comprised between the NE-trending Cévennes and Nîmes faults during the Priabonian (Séranne et al., 2021). The elevated sedimentation rates, greater than 200 m/Myr, revealed in the Vistrenque Basin are consistent with a basin subsidence controlled by the major Nîmes strike-slip fault. In contrast with Languedoc basins where early to middle Rupelian are lacking, the sedimentation in the Vistrenque Basin suggests that the strike-slip movement of the Nîmes

fault was still active during the Rupelian. In addition, the sharp decrease in sedimentation rates between the upper part of the *Série Grise* (31.2 Ma in Gallician-9) and the *Série Rouge* (27.88 Ma in Gallician-9) suggest that the strike-slip movement of the Nîmes fault, associated to the ongoing shortening in the Spanish Pyrénées, ceased in the middle to late Rupelian times. The *Série Grise* Formation from the Vistrenque Basin records therefore the latest stages of Pyrenean compression until the Rupelian together with the earliest stages of the dismantling of the Languedoc-Provence segment of Pyrénées. Finally, the Chattian evaporitic succession (*Série Calcareo-salifère* Formation) and the overlying Aquitanian terrigenous sequence were deposited during the Liguro-Provençal Rifting stage (Benedicto, 1996; Séranne et al., 2021).

5.3 Evidence of marine incursions into Priabonian to Chattian continental basins from

Southeast France

Saline lake sedimentation associated with the deposition of sulfates (gypsum and anhydrite) occurred during the Priabonian and/or Rupelian in various basins from SE France: Alès, Saint-Chaptes and Issirac basins (Lettéron et al., 2017, 2018, 2022) as well as in the Mormoiron-Carpentras and Apt-Manosque-Forcalquier basins (e.g. Triat and Truc, 1974; Cavelier et al., 1984). The present work documents saline lake conditions and deposition of evaporites in the Vistrenque Basin as early as the early Rupelian (within the *Série Grise*) and during the Chattian (*Série Calcaréo-salifère*).

The origin of dissolved ions (Briot, 2008) in the Paleogene saline lake waters, and the possibilities and pathways of salinization of these lacustrine systems of Southeast France and Western-European rift system are still discussed (e.g., Lettéron et al., 2017, 2018, 2022; Semmani et al., 2022). Lettéron et al. (2017) considered the relevance of the occurrence of benthic foraminifera and halophile molluscs in the lacustrine deposits from the Issirac Basin to demonstrate marine connections and discarded evidence of marine connections based on these

faunal contents. Moreover, these organisms have been shown to be tolerant to salinity variations and can thrive in athalassic lakes (Plaziat and Younis, 2005). Nevertheless, strong evidence of incursions of marine-influenced waters within the ASCI lake system during the Priabonian and the Vistrenque Basin from the Late Eocene to Late Oligocene can be provided by the occurrence of calcareous nannofossils, dinoflagellate cysts and halophilous plant pollen grains, and by the contribution of geochemical modelling.

In the ASCI system, the occurrence of marine dinoflagellate cysts in Serre-de-Cauvel and at the crossroad D716/D981 (Saint-Chaptes Basin: Fig. 4) is indicative of marine connections as early as the early-to-middle Priabonian. Additionally, the presence of *Avicennia* pollen grains in the Saint-Chaptes (Crossroads D716/D981) and Alès basins (Célas Tunnel, Saint-Jean de Maruéjols Pit hill and Montchamp) suggests the persistent influence of marine water from the middle to the latest Priabonian. The occurrence of Rhizophoraceae pollen grains from Serre de Cauvel and Montchamp is consistent with such an interpretation. Finally, the Sr and S isotope signatures of evaporites from the Ales Basin, although altered by sulfate-reduction processes likely derive from a mixture between a continental water influenced by Triassic evaporites and seawater.

In the Vistrenque Basin, the presence of the dinoflagellate *Deflandrea phosphoritica* (chronological range: 54.6-21.2 Ma; Powell et al., 1996) in the *Série Grise* (Gallician 9 well at 2530 m depth) provides evidence of marine incursion into the Vistrenque Basin during the early Rupelian times. The presence of pollen grains of *Ephedra* is a characteristic feature of this pollen record. The present-day species of *Ephedra* are all living in arid and semi-arid regions (González-Juárez et al., 2020). The few species growing on coastal sands are not salt adapted and thus are not considered as halophytes (Halophytes Database). For example, *Ephedra*, which is currently frequent along the Mediterranean coastline (*E. distachya* L.: Coste, 1937), is represented by very scarce pollen grains in present-day pollen assemblages from the Camargue

and Roussillon coastal areas (Cambon et al., 1997; Beaudouin et al., 2005a; Suc and Fauquette, 2012). It is also very scarce in the pollen records of warm phases (Holocene and older interglacials) although it is frequent to abundant during the drier episodes corresponding to cold phases (Beaudouin et al., 2005b; Tesson et al., 2011; Arnaud-Fassetta and Suc, 2015). This comparison with modern to recent pollen records supports the interpretation of *Ephedra* as marker of both cooling and aridification phases rather than of marine incursion. On the other hand, the Sr and S isotope signatures of sulfate evaporites from the *Série Calcaréo-salifère* Formation and mixing-water modelling strongly suggest an intrusion of marine water into the Vistrenque Basin during the late Chattian.

5.4 Paleogeographic reconstructions of Southeast France during the Late Eocene and

Oligocene

The potential pathways of the marine incursions into the Southeast France continental basin are displayed in the palaeogeographic reconstructions of Figures 11, 12, and 13. During the Priabonian and Rupelian, the Southeast France lake basins as well as the West Alpine Sea were bounded to the south by the EW-trending Pyrenean-Corsica-Sardinia Massif (Joseph and Lomas, 2004; Séranne et al., 2021). This relief likely yielded most of the terrigenous supply of the ASCI and Vistrenque lake basins (Lettéron et al., 2022; Semmani et al., 2022) and deepmarine West Alpine foreland Basin (Joseph and Lomas, 2004). Moreover, it was likely sufficiently continuous and elevated to prevent from connections with the Western Neotethys to the south (Fig. 11 and Fig. 12). In addition, the Atlantic shoreline was located at more than 300 km west of the Alès Basin during the Priabonian and Rupelian times (Gely and Sztrákos, 2000; Sztrákos and Steurbaut, 2017), and fluvial sedimentation dominates in south-western Languedoc basins (Calvet et al., 2021; Séranne et al., 2021) thus making unlikely marine intrusion from the Atlantic Ocean into Languedoc, Rhône Valley and Camargue areas. Finally,

the demise of lacustrine sedimentation in the ASCI basins during the latest Priabonian or earliest Rupelian and the depositional hiatus of most of the Rupelian interval (Lettéron et al., 2022) do not support connections with the marine realm west of the Nîmes fault during the Rupelian (Fig. 12).

In contrast, the proximity of the West Alpine foreland (<30 km from the Manosque-Forcalquier Basin, and <50 km from the Valence Basin), and the probable interconnection between the different SE France saline lake basins, as suggested by previous studies (Lettéron et al., 2022; Semmani et al., 2022) makes marine intrusions more plausible from the West Alpine Sea during the Priabonian and Rupelian (Fig. 11 and Fig. 12).

The reconstruction of Pyrenean structures and their differentiation from later Alpine deformation (Balansa et al., 2022) provides a structural framework allowing a realistic reconstruction of continental paleo-reliefs during the Priabonian and Rupelian and the identification of two potential marine incursion pathways, from the West Alpine Sea to the Southeast France lake basins (Fig. 11 and Fig. 12). A northern potential pathway would connect the West Alpine Sea with the Valence Graben, north of the Diois-Barronies High. This corresponds with the "Crest Sill" identified by Sissingh (2001) as a possible marine corridor supplying seawater into the Western European rift basins. A connection of the Valence lake Basin with the Mormoiron and Vistrenque to the south and finally with the ASCI system during periods of maximum lake expansion would therefore explain the occurrence of calcareous nannofossils, dinoflagellate cysts and mangrove pollen grains within the Alès Basin during the Priabonian and in the Vistrenque Basin during the Rupelian.

A southern potential pathway of marine incursion would connect the West Alpine Sea with the Manosque-Forcalquier, Mormoiron and Vistrenque-ASCI basins through a set of Pyrenean synclines in the Haut-Var region (Bauduen, Montmeyan, Salernes and Rians basins). The hypothesis of such a marine corridor is corroborated by the presence of Paleogene marine markers such as calcareous nannofossils and dinoflagellate cysts in the "Sables Bleutés du Var" Formation in the Montmeyan Basin (Châteauneuf et al., 2019) even though the age of these deposits remains very much debated (e.g., Philip et al., 2017). Such a southern sea-water pathway may also explain the occurrence of dinoflagellate cysts in the Rupelian from the Marseilles Basin (Châteuneuf and Nury, 1995).

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The Chattian paleography of SE France (Fig. 13) differs significantly from that of Priabonian-Rupelian times, since the West Alpine Sea closed as a result of sedimentary infill and westward accretion of the Alpine orogenic wedge (Joseph and Lomas, 2004) during the late Rupelian. In addition, the Chattian is regionally characterized by widespread active rifting that formed NE-SW oriented rift-basins, onshore and offshore in the Gulf of Lion. This rifting led to the collapse of the Pyrenean-Corsica-Sardinia. This resulted in marine intrusions from the southern Western Neotethys, into Provence as evidenced by the occurrence of late Chattian, strictly marine, shallow-water carbonate sediments west of Marseilles ('Côte Bleue' shoreline: Oudet et al., 2010a) and marine biological markers including dinoflagellate cysts, coelenterates and marine fishes in brackish-water lacustrine/lagoonal deposits from the Aix-en-Provence Basin (Nury and Thomassin, 1994). The marine influence revealed by strontium and sulfur isotope signatures of Chattian evaporites from the Vistrengue Basin could therefore results from a marine connection to the south probably through the offshore Vistrengue Basin, the Central Graben and the Catalan fault zone domain (Fig. 13). Such a marine water pathway, suggested by Oudet et al. (2010b) is supported by the presence of Chattian to Aquitanian planktonic foraminifera in well Tramontane 1 in the Central Graben (Cravatte et al., 1974). The Vistrenque Basin may also have been connected with the Sardinian Graben (via Offshore Vistrenque and Grand Faraman Graben) where the earliest marine ingression occurred during the latest Chattian-early Aquitanian (e.g. Casula et al., 2001). The Chattian carbonate and evaporite sediments from the Série Calcaréo-salifère Formation in the Vistrenque Basin could therefore

have deposited in a saline lake with transient connections with seawater during sea-level or lake-level highstands, or within a coastal sabkha with periodic marine intrusions.

5.5 Paleoclimatic implications

This present study, based on both botanical pollen identification and counting, robustly documents for the first time the climate evolution of the 38–23 Ma interval in South of France, which encompasses the Eocene–Oligocene transition. The four pollen floras from the Canet 1 borehole complete those from the Alès and Vistrenque basins for documenting the climate knowledge during the late Rupelian and early Chattian.

According to our pollen data, we demonstrate that the Eocene–Oligocene transition is marked by the suddenness and the great amplitude of the climate deterioration with lowering temperature and increasing dryness at low elevations while high humidity continued in higher altitude. The climate parameters estimated from analyses of pollen from the Vistrenque samples do not show a drastic climate deterioration. This may be due to the large climatic tolerance intervals attributed to the plants, some of them may live under large temperature and/or precipitation amplitudes at the genus or family level. However, the cooling between the Priabonian and the Rupelian is indicated by a decrease in mean annual temperature, mean temperature of the coldest month and in mean annual precipitation at Vauvert but more particularly at Gallician 9 2530 m. Following that event, temperature and precipitation increase, although they remain lower than during the Priabonian. Vegetation and climate changes, deduced from the relative abundance of megathermal and mega-mesothermal vs. herbaceous and steppic plants in the pollen diagrams, seem to indicate that the cooling in Southeast France was brief and took place at the very early Rupelian. From a vegetation and climatic point of view, the Mediterranean region is considered as a very particular region since the Paleogene as

it was certainly sheltered from cold air masses (Popescu et al., 2021). The cooling phases were thus certainly less pronounced than elsewhere in Europe.

Finally, the upper Chattian succession shows a climate evolution towards the Aquitanian conditions with very scarce megathermal plants as described by Bessedik (1984), Zheng (1990) and Suc et al. (2018), despite persistence of *Podocarpus* in the mountains up to the latest Chattian.

6. Conclusion

- The definition of a robust chronostratigraphic framework for the Vistrenque Basin and the neighboring Paleogene basins, combined with the analysis of a set of micropaleontological and geochemical data, has led to significant improvements in knowledge of the geodynamic evolution, the paleogeographic framework and the paleoclimatic setting of the Upper Eocene-Oligocene lacustrine basins from South-East France:
 - 1) The *Série Grise* Formation (Priabonian to early Rupelian) from the Vistrenque Basin was deposited in a highly subsident basin associated with the strike-slip movement of the Nîmes fault and records the latest stages of the Pyrenean shortening. Drastic decrease in sedimentation rates above the *Série Grise* suggests that the strike-slip movement of the Nîmes fault ceased around the middle-to late Rupelian. The evaporitic sedimentation of the *Série Calcareo-salifère* Formation (Chattian) is related to the stage of Liguro-Provençal rifting.
 - 2) The pollen data recorded during this study allow climatostratigraphic correlations with the reference oxygen isotope curve and thus help to specify the age of the studied sections. The floral assemblages from the Priabonian and Rupelian of the neighbouring

Languedoc and Camargue areas constitute a powerful tool to document the climatic deterioration at the Eocene-Oligocene transition. In addition, pollen analyses made it possible to reconstruct a mangrove and freshwater lake vegetation in the Alès Basin, and a dominant evergreen subtropical forest in the surroundings of the lake which also record fluctuations with a high-altitude vegetation in the southern paleo-Pyrenean relief during the Priabonian. Vegetation cover has been shown to be rather dominated by mesothermal and herbaceous plants. During the early Rupelian and Chattian times, a clear influence of altitude vegetation is referred to the inherited Pyrenean relief, south of the study area. Simultaneously, cooler and drier climate conditions affected the surroundings of the Camargue area.

- 3) The use of the Climate Amplitude Method allowed evidencing a short but significant stage of climate cooling and aridification in the early Rupelian which could result from the global climate cooling following the Eocene-Oligocene Transition.
- 4) Strong evidence for successive intrusions of marine-influenced saltwater into the Languedoc and Camargue areas during the Priabonian, early Rupelian and Chattian intervals is provided by the occurrence of calcareous nannofossils, dinoflagellate cysts and the halophilous Avicennia and Rhizophoracea plants documented by their pollen. Strontium and sulfur isotope signatures of sulfates from the Série Calcaréo-salifère Formation strongly suggest incursions of marine water into the Vistrenque lacustrine system during the Chattian.
- 5) During the Priabonian and Rupelian, marine water likely derived from the Western Alpine Sea. Two possible pathways of seawater intrusion are suggested from paleogeographic reconstructions: the Crest Sill north to the Diois-Barronies High and/or the Pyrenean synclines in the Haut-Var area south of the Diois-Barronies High. The paleogeographic setting of south-east France significantly changed as a result of the

closure of the Alpine Sea and the formation of rift basins in the Gulf of Lion during the Chattian have made possible marine incursion into the Vistrenque Basin from the Mediterranean domain to the south.

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Figure and table caption

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Figure 1: A) Paleotectonic map of Western Europe during the Oligocene-Aquitanian times showing the location of Western European rift basins (ECRIS) and the subduction area to the south and southeast of Corsica-Sardinia, from Bois (1993). B) Structural map of Southeast France and location of the Vistrengue Basin among the various Cenozoic continental basins, modified after Benedicto (1996). Sampling loci for pollen analyses, U/Pb dating, and Sulfur/Strontium isotopes purposes are located in the Alès, Issirac and Saint-Chaptes basins (AB, IB, SCB), Vistrenque Graben (VG) and Roussillon Basin (RB), geographic coordinates are given in Supplementary data (Table S1). C) Geological section across the Gulf of Lion margin (modified after Benedicto, 1996). Figure 2: A) Map of the Bouguer gravity anomaly, modified after Chauvin and Follin-Arbelet (1985). B) interpreted seismic profile SG3 in the Northern Vauvert compartment, after Benedicto (1996), (see location in Fig. 3 hereinafter). C) NW-SE geological cross-section through the Vistrenque Basin on the northern Vauvert compartment (modified after Benedicto et al., 1996). D) Stratigraphic synthesis of the Vistrengue Basin compiled from the boreholes data by Gorini (1993), modified after Bache (2008). Figure 3: Vistrenque Basin: A) Lithological columns and correlations of Gallician 9 and Pierrefeu 1 wells (after Semmani, 2022); B) Structural map of the Vistrenque Basin and well location (after Benedicto et al., 1996); C) Location map of wells and seismic profiles from the

Gallician and Pierrefeu area (location: black rectangle on Fig. 3B).

Figure 4: A) Geological map of the Alès-Saint-Chaptes-Issirac (ASCI) basins and location of the pollen sampling. B) Chronostratigraphic chart of the ASCI lake system (after Lettéron et al., 2022) and location of palynological and sulfate samples analyzed in this study.

Figure 5: Correlation panel of wells Gallician 1, Gallician 4, Gallician 9, Vauvert 1 and Pierrefeu 1 from the Vistrenque Basin (after Semmani, 2022) showing the sedimentary units, lithologies and lithostratigraphic correlations. Lithostratigraphic correlations are carried out using lithological similarities and gamma ray (GR) signal. Drill core locations and the positions of samples used for U/Pb dating (squares) and pollen analyses (circles), and 87 Sr/ 86 Sr & δ^{34} S measurements (triangles) are also plotted.

Figure 6: A) Microphotograph of the cavity in thin section of the sample G9 13D from Gallician 9 well (depth ~ 1960 m) displaying the growth of successive cement stages: microstalactitic cements A1 & A2, and coarse bladed calcite I1 with further bitumen infill. B) elemental mapping of the red box in A) showing the concentrations of ²³⁸U and ²⁰⁶P. C) Confocal microscopy photograph of the cavity in (A) where the fluorescence allows to observe the different generations of cement. The cementation stages A2 and I1 are non-luminescent and therefore indistinguishable under cathodoluminescence. D) Tera-Wasserburg plots displaying ²³⁸U/²⁰⁶Pb versus ²⁰⁷Pb/²⁰⁶Pb from the A1 cement (early diagenetic anisopachous cement).

Figure 7: A) Thick geological section scan (sample G9 7; Gallician 9 well, depth ~ 1894 m) showing the yellowish beige pendant anisopachous cement around clasts in a fluvial conglomerate. B) close-up view of the rectangular box in A) displaying the position of the laser ablation spots alongside the cement. C) Tera-Wasserburg plots displaying ²³⁸U/²⁰⁶Pb versus ²⁰⁷Pb/²⁰⁶Pb from the anisopachous cement of the G9 7 sample (number of spots = 74). D) and E) Microphotographs of the vein calcite infill within the G9 K35 sample (Gallician 9, depth ~ 2487 m) under plane polarized light and cathodoluminescence respectively. Note the dull

luminescence of the calcite fill (right hand picture). F) Tera-Wasserburg plots displaying 1340 ²³⁸U/²⁰⁶Pb versus ²⁰⁷Pb/²⁰⁶Pb from the calcite vein infill of the G9 K35 sample (number of spots 1341 = 45).

Figure 8: Synthetic pollen diagrams (*Pinus* excluded) illustrating (1) to the left the local to regional paleovegetation (including altitudinal trees) and (2) to the right the local paaeovegetation (the latter trees excluded) – their proposed climatostratigraphic correlations with the reference oxygen isotope curve (smoothed curve over 20 ka in blue, smoothed curve over 1 Ma in red; Westerhold et al., 2020). Pollen diagrams are vertically arranged according to the basin stratigraphy and available chronostratigraphic information (mammal faunas, calcareous nannofossils). Time-intervals corresponding to the Priabonian and Chattian stages are shaded in light grey to make easier the chronostratigraphic correlations. Pollen group composition is specified in the text and more in **Supplementary data Table S3**.

Figure 9: δ^{34} S vs 87 Sr/ 86 Sr crossplot for Paleogene anhydrites from SE France: A) Vistrenque Basin (Pierrefeu 1 well); the solid line represents the water-mixing model for a mixture of a Late Oligocene seawater with a riverine water whose composition is influenced by recycling of Triassic evaporites (see **Table 3** for Sr and S isotope compositions and concentrations of each end-member); the dashed area represents possible isotope compositions of sulfates having precipitated from the modelled water mixture by considering the fractional crystallization effect; B) Alès Basin (Les Fumades: SC-1 and SC-2 wells). The arrow indicates how bacterial sulfate reduction results into higher δ^{34} S. The Triassic (Keuper) evaporite domain is compiled from Albarède and Michard (1987), Dinelli et al. (1999), Alonso-Azcarate *et al.* (2006), Ortí *et al.* (2014), Utrilla *et al.* (1992) and Boschetti *et al.* (2011). The granite and metamorphic rock domain derives from Petelet-Giraud *et al.* (2006), Chaussidon *et al.* (1989), Giacometti et al. (2014).

1364	Figure 10: Chronostratigraphic framework of the Paleogene succession of the Vistrenque
1365	Basin. Stratigraphic attributions for the main lithological units are obtained using U/Pb absolute
1366	dating (blue squares), pollen analyses and geochemical modelling on sulfates, see text for
1367	details about the dating results. Reference oxygen isotope curve (smoothed curve over 20 ka in
1368	blue, smoothed curve over 1 Ma in red; Westerhold et al., 2020). Climatic events and Antarctic
1369	ice-sheets coverages are derived from Zachos et al. (2001).
1370	Figure 11: Paleogeographic map of Southeast France during the late Priabonian. Location of
1371	continental basins are compiled from Lettéron et al. (2017, 2022) and Semmani et al. (2022).
1372	Paleogeography of marine domain is modified from Joseph and Lomas (2004). The structural
1373	framework is compiled from Séranne et al. (2021) and Semmani et al. (2022) west of the Diois-
1374	Barronies High and from Balansa et al. (2022) for Northern and Eastern Provence.
1375	Figure 12: Paleogeographic map of Southeast France during the early-middle Rupelian.
1376	Location of continental basins are compiled from Nury (1990), Lettéron et al. (2017, 2022),
1377	Semmani et al. (2022). Paleogeography of marine domain is modified from Joseph and Lomas
1378	(2004). The structural framework is compiled from Séranne et al. (2021) and Semmani et al.
1379	(2022) west of the Diois-Barronies High and from Balansa et al. (2022) for Northern and
1380	Eastern Provence.
1381	Figure 13: Paleogeographic map of South-East France during the Chattian. Location of
1382	continental basins are compiled from Cavelier et al. (1984), Nury (1990), Séranne et al. (2021).
1383	Paleogeography of marine domain is modified from Oudet (2008).
1384	

1387	
1388	Table 1: Ages obtained in the Vistrenque Basin using U/Pb dating on calcite, pollen-based
1389	climatostratigraphy, Sr and S isotope signatures of sulfates.
1390	Table 2: Climatic reconstruction based on pollen data. The succession of pollen sites is that
1391	shown in Figure 8. Mean annual temperatures (MAT), mean temperature of the coldest (MTC)
1392	and of the warmest (MTW) months and mean annual precipitation (MAP) are reconstructed
1393	using the Climatic Amplitude Method (Fauquette et al., 1998). The results are given as an
1394	interval and a Most Likely Value (in bold) for each site.
1395	Table 3 : 87 Sr/ 86 Sr and δ^{34} S measurements on anhydrite/gypsum samples and native sulfur from
	Table 5. St. St and 6. Still easurements on annyurite/gypsum samples and native surfur from
1396	Pierrefeu-1 well (Vistrenque Basin), SC-1 and SC-2 well (Alès Basin). The stratigraphic
1396	Pierrefeu-1 well (Vistrenque Basin), SC-1 and SC-2 well (Alès Basin). The stratigraphic
1396	Pierrefeu-1 well (Vistrenque Basin), SC-1 and SC-2 well (Alès Basin). The stratigraphic
1396 1397	Pierrefeu-1 well (Vistrenque Basin), SC-1 and SC-2 well (Alès Basin). The stratigraphic

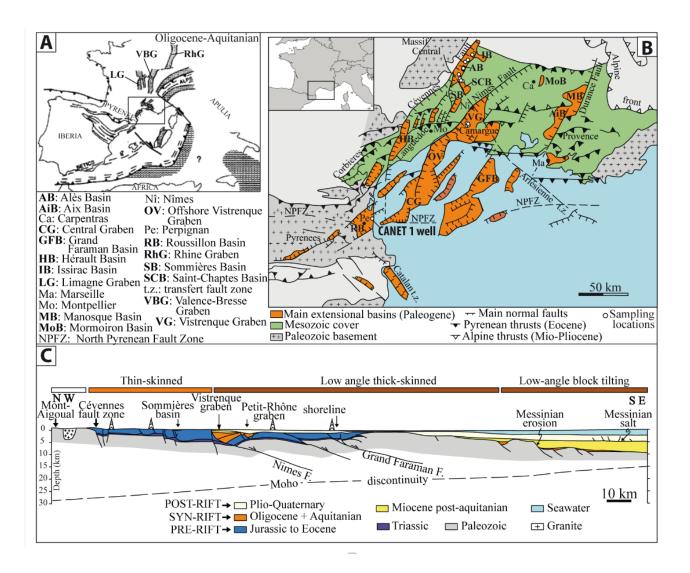


Figure 1

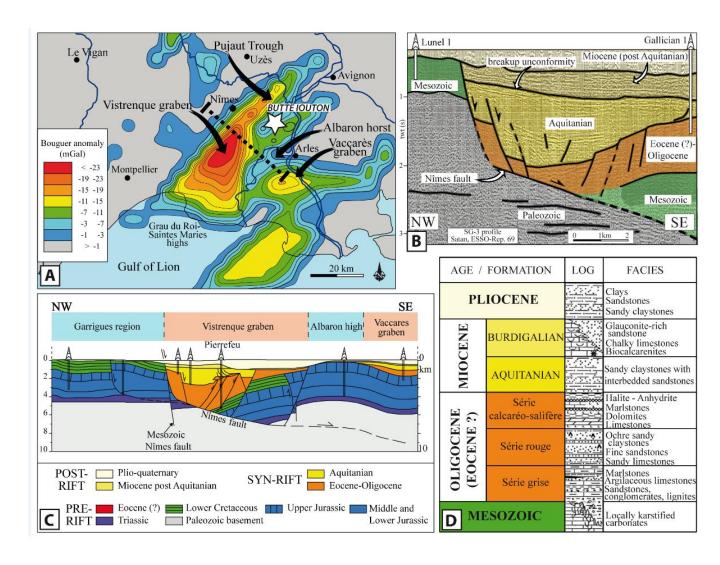


Figure 2

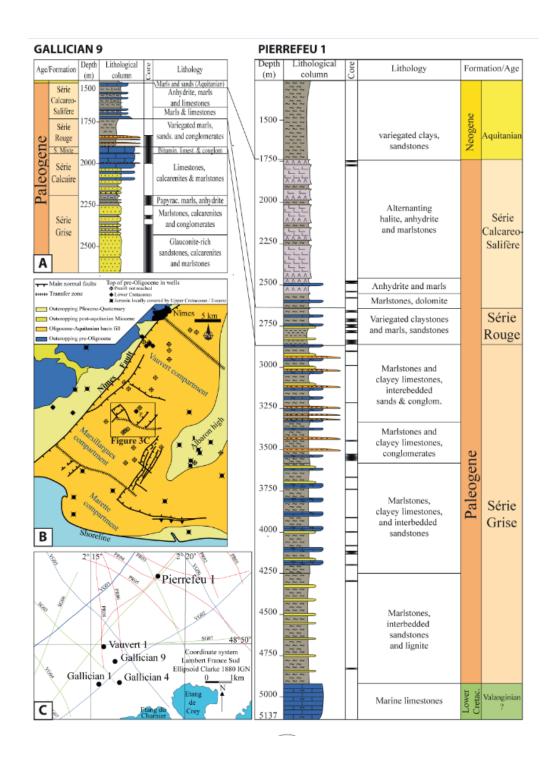


Figure 3

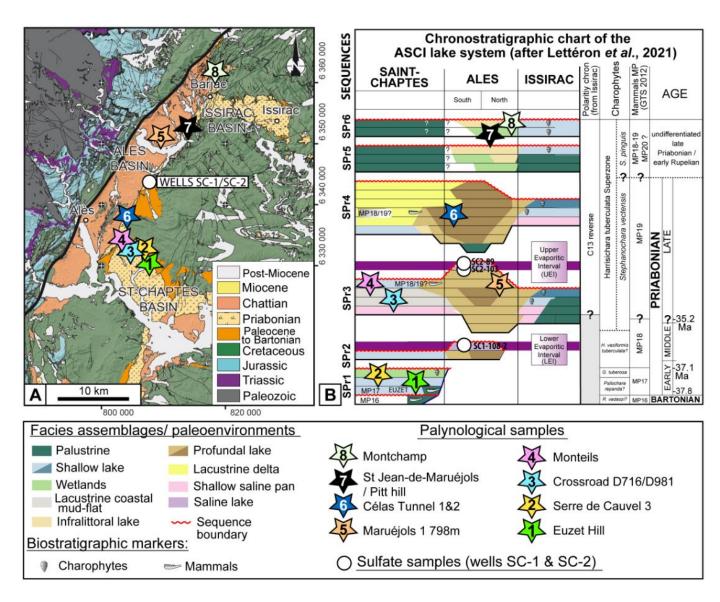


Figure 4

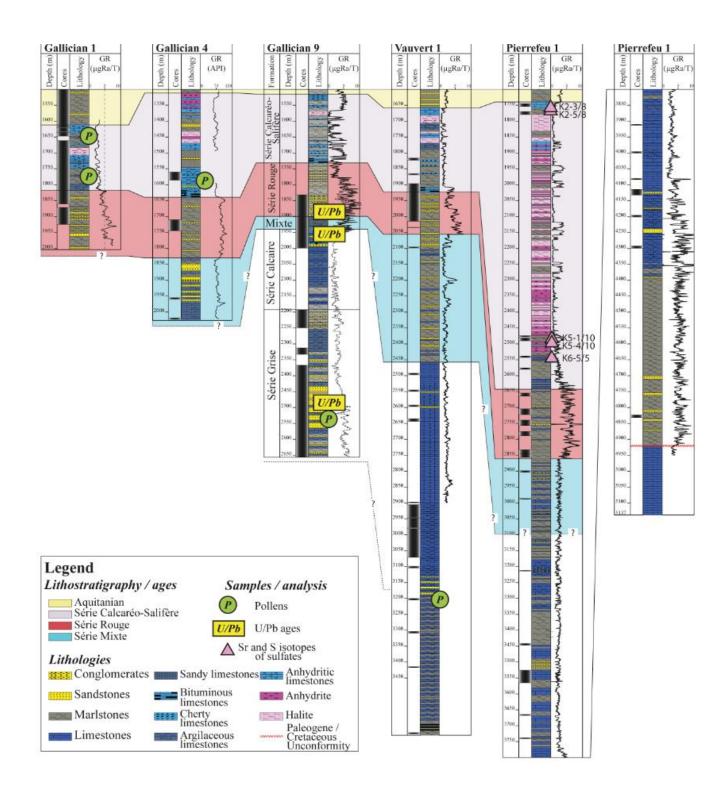


Figure 5

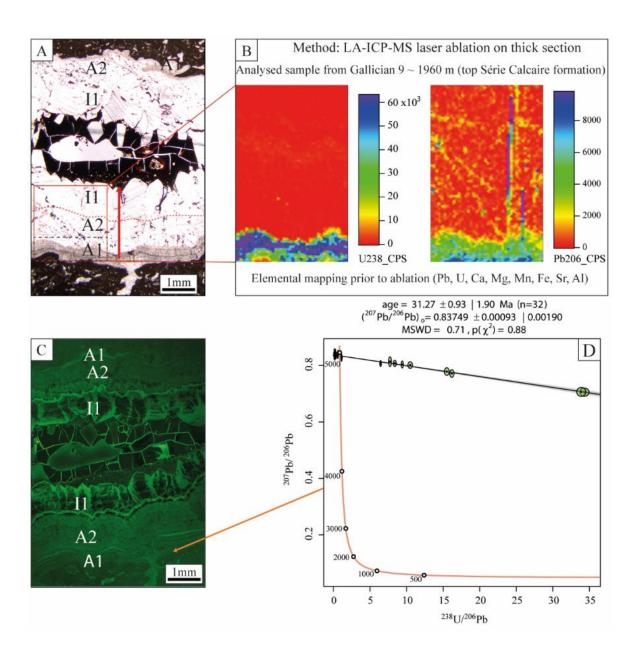


Figure 6

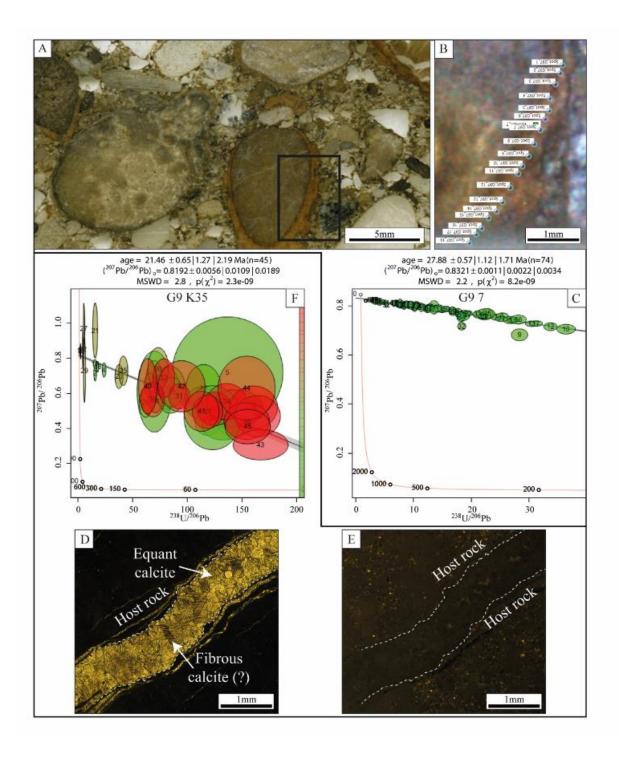
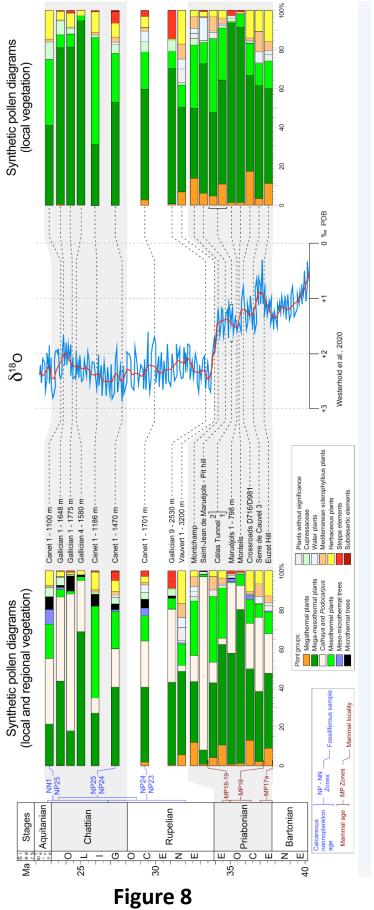


Figure 7



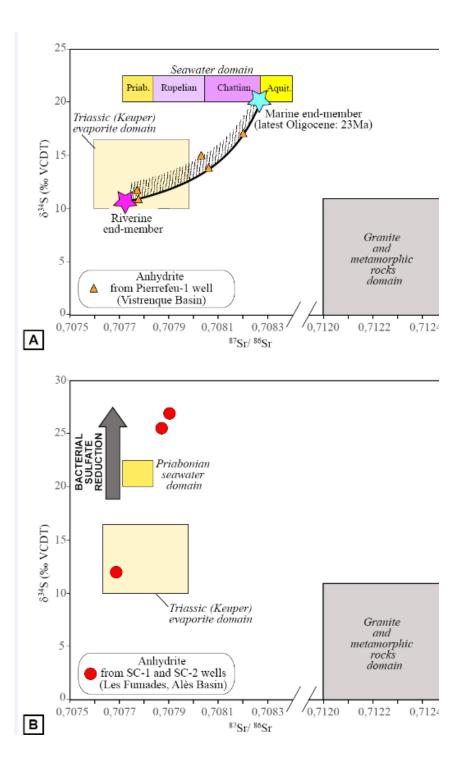


Figure 9

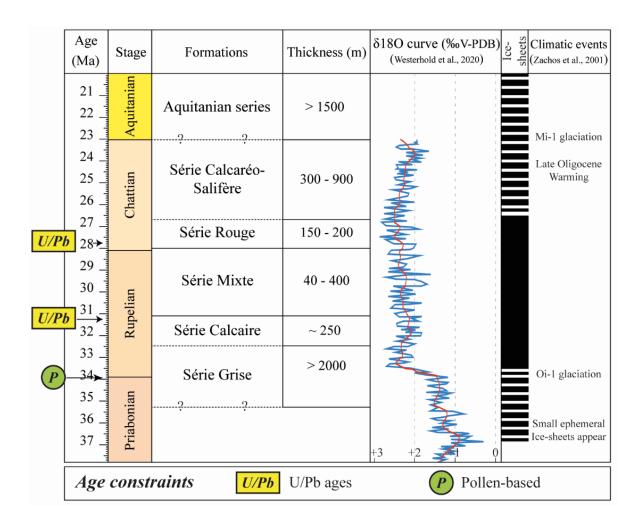


Figure 10

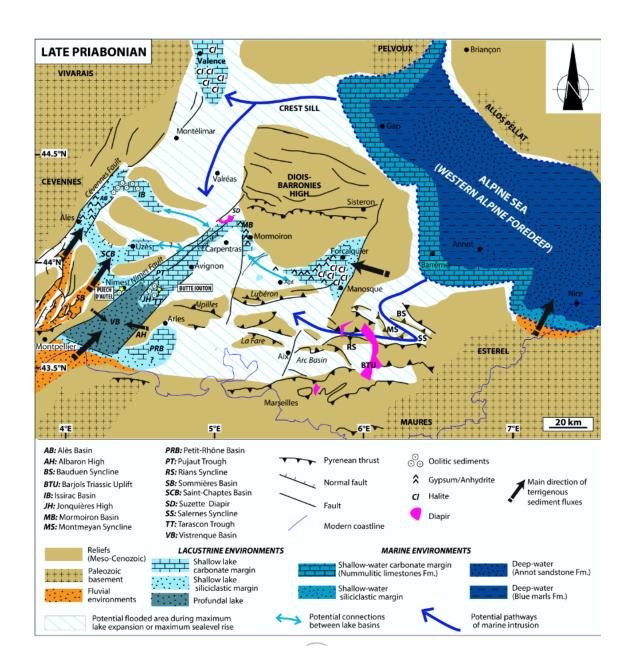


Figure 11

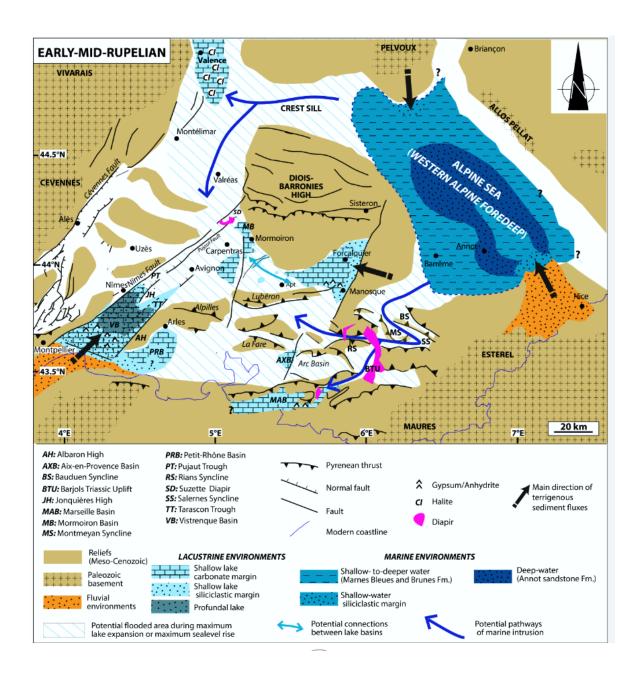


Figure 12

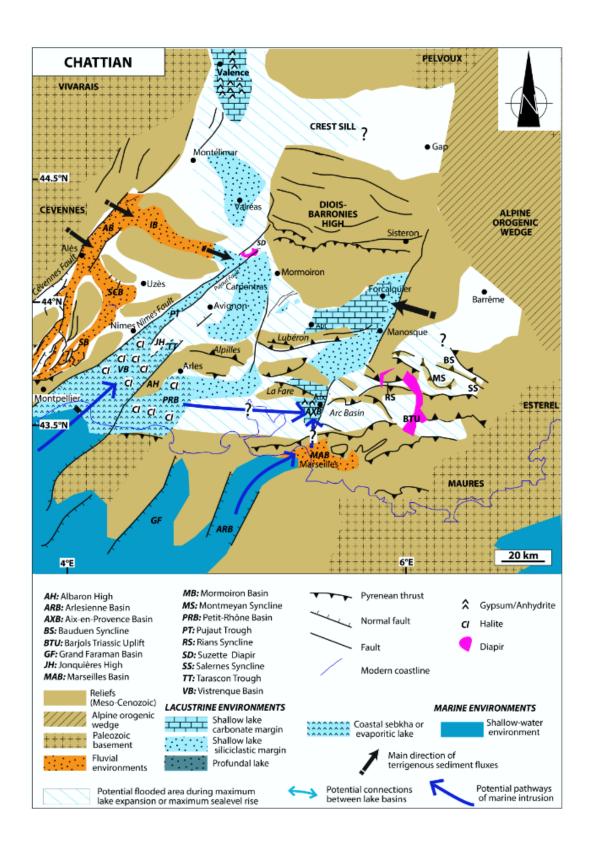


Figure 13

Lithostratigraphic units	Age	Dating approach	Analysed sample	Well - depth(m)
Série Calcaréo- salifère	Late Chattian to earliest Aquitanian?	Geochemical modelling on sulphates	Anhydrite	Pierrefeu 1 – 1750 1775 – 2485 – 2545
	Early to middle Chattian	Pollen <u>climatostratigrpahy</u>	Pollen assemblage	Gallician 1 – 1648 Gallician 1 – 1775 Gallician 4 – 1580
Série Rouge	Early Chattian (a)	U/Pb dating * Absolute age: 27.88 +/1.71 Ma (b)	Vadose cement	Gallician 9 – 1894
Série <u>Mixte</u>	Middle Rupelian to early Chattian	Bounding	/	/
Série <u>Calcaire</u>	Early to middle Rupelian (a)	U/Pb dating * Absolute age: 31.27 +/1.90 Ma (b)	Vadose cement	Gallician 9 – 1960
Série Grise	Priabonian (?) to early Rupelian	Pollen <u>climatostratigraphy</u>	Pollen assemblage	Gallician 9 – 2530 Vauvert 1 – 3200
	(b) Ages are calibrated with the Ge	cological time Scale after Gradstein et al. (2012) rrors and errors associated with statistic calcula		

Site	Basin	MAT (°C)	MTC (°C)	MTW (°C)	MAP (mm)
Canet 1 - 1100 m	Roussillon	16 -19- 25	10 -11.6- 14	25 -26.2- 29	1100 -1640- 2200
Gallician 1 - 1648 m	Vistrenque	16- 18.9 -25	5-9- 14	25- 26.2 -29	1100- 1560 -1960
Canet 1 - 1186 m	Roussillon	15- 18.4 -25	2.5- 10.4 -14	25- 26.2 -29	1000- 1260 -1400
Canet 1 - 1470 m	Roussillon	16- 19 .1-24	10- 11.7 -14	25- 26.1 -28	1100- 1230 -1300
Canet 1 - 1701 m	Roussillon	19- 20.7 -25	10- 12.7 -16.5	26- 26.7 -28	1200- 1530 -1960
Gallician 9 - 2530 m	Vistrenque	12- 15.8 -20	2.5- 7.3 -14	20- 23.3 -27	800- 870 -2200
Vauvert 1 - 3200 m	Vistrenque	17- 19.7 -25	12- 13.2 -15	25- 26.2 -28	1200- 1325 -1400
Montchamp	Alès	20- 21.4 -25	12- 14.4 -15	26- 26.4 -28	1100- 1530 -1800
Celas Tunnel 2	Alès	19- 20.9 -25	12- 14.5 -17	25- 26.7 -29	1500- 1670 -1800
Celas Tunnel 1	Alès	19- 20.9 -25	10- 12.5 -15	26- 26.9 -28	1500- 1760 -2000
Maruéjols 1 - 798 m	Alès	19- 20.9 -25	12-14-17	26- 27.1 -29	1100 -1630 -2000
Monteils	Saint- Chaptes	15- 18.7 -25	10- 11.7 -14	25- 26.1 -28	1500- 1650 -1800
Crossroads	Saint-	19- 21.1 -25	12 -13 -14	26- 27 -29	1000- 1400 -1560
D716/D981	Chaptes				
Serre de Cauvel 3	Saint- Chaptes	18- 20.2 -25	12- 12.8 -14	25 -26.2 -28	1100 -1540 -1800
Euzet Hill	Saint- Chaptes	17- 20.1 -24	12- 13.9 -16	25 -26.3 -28	1200- 1530 -1600

Well	Formation/ Sequence	Sample ID	Lithology	Age	⁸⁷ Sr/ ⁸⁶ Sr	δ ³⁴ S ‰VCDT
	Calcaréo- salifère	K2-3/8	anhydrite	Oligocene	0,707777	+10,92
	Calcaréo- salifère	K2-5/8	anhydrite	Oligocene	0,707772	+11,75
Pierrefeu 1 (Vistrenque Basin)	Calcaréo- salifère	K5-1/10	anhydrite	Oligocene	0,708061	+13,87
	Calcaréo- salifère	K5-4/10	anhydrite	Oligocene	0,708031	+15,02
	Calcaréo- salifère	K6-5/5	anhydrite	Oligocene	0,708201	+17,11
	Sequence SPr2	SC1-108-2	anhydrite/gypsum	Priabonian	0,707685	+12,11
SC-1 well (Alès	Sequence SPr3	SC2-103	anhydrite/gypsum	Priabonian	0,707903	+26,98
Basin) SC-2 well (Alès Basin)	Sequence SPr3	SC2-89	anhydrite/gypsum	Priabonian	0,707870	+25,59
	Sequence SPr3	SC2-89	native sulphur	Priabonian		-6.42
	Sequence SPr3	SC2-89	native sulphur	Priabonian		-5.99

SUPPLEMENTARY DATA

MICROPALEONTOLOGY: CALCAREOUS NANNOFOSSILS AND PALYNOLOGY

The micropalaeontological content (pollen grains and spores, dinoflagellate cysts, calcareous nannofossils) of thirteen locations (boreholes and exposed sections) has been searched. They belong to the Vistrenque Basin (Gallician 1 – 4 – 9, Vauvert 1), the Alès Basin (Maruéjols 1, Euzet Hill, Serre de Cauvel, Crossroad D716/D981, Monteils, Célas Tunnel, Saint-Jean de Maruéjols Pit hill, Montchamp) and the Roussillon Basin (Canet 1) (Fig. Y; Table S1). Occurrences of calcareous nannofossils are presented in Table S2, palynological results are detailed in Table S3.

Location	North Latitude	East Longitude	Elevation (m)	Basin
Canet 1	42°41'23.41"	3°04'66.00"	-1099 -1700	Roussillon
Gallician 9	43°38'35.64"	4°16'58.49"	-2515	
Gallician 4	43°38'13.86"	4°17'06.36"	-1581	Vistrenque
Gallician 1	43°38'14.98"	4°16'58.22"	-1647 -1775	
Vauvert 1	43°38'58.04"	4°16'39.43"	-3177	
Euzet Hill	44°04'35.71"	4°12'41.02"	127	
Serre de Cauvel	44°04'36.90"	4°12'28.10"	120	
Crossroad D716/D981	44°04'50.50"	4°11'57.50"	132	
Monteils	44°05'08.00"	4°11'39.00"	168	Alès
Célas Tunnel	44°07'37.70"	4°10'34.80"	201	
Saint-Jean de Maruejols Pit hill	44°15'59.05"	4°17'48.36"	128	
Maruéjols 1	44°15'35.76"	4°15'20.78"	-633	
Montchamp	44°19'56.20"	4°21'29.30"	220	

Table S1. Latitude, longitude, elevation and reference basin of the studied locations.

For boreholes, elevation refers to depth of samples with respect to elevation of the hole top.

Canet 1 borehole.

Six samples were chosen within clay layers from the lithostratigraphic interval ascribed to Aquitanian–late Oligocene by Gottis (1958) in order to obtain pollen grains, four of them revealed a rich pollen flora (Fig. S1) and three of them contained abundant dinoflagellate cysts. As a consequence, calcareous nannofossils were successfully searched in these last samples (Table S2).

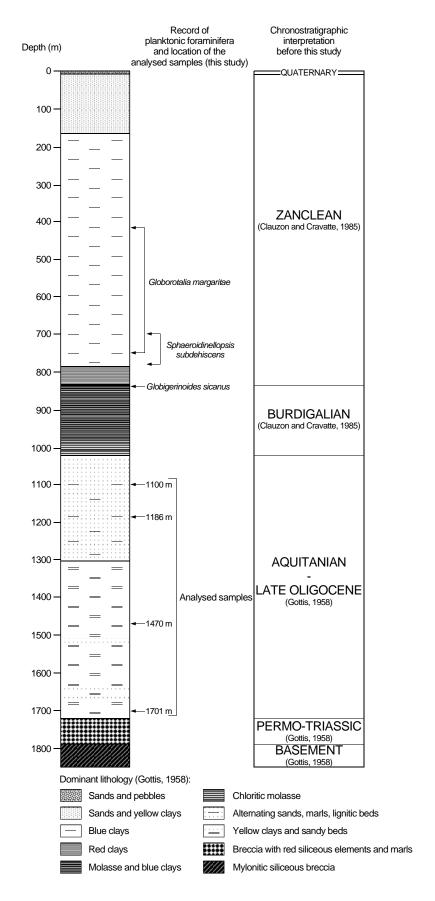


Figure S1. Canet 1 borehole: lithostratigraphic log, biostratigraphic markers, location of the analysed samples in this study, chronostratigraphic interpretation previous to this study.

Samples	Calcareous nannofossils
Gallician 1 – 1594 m	Coccolithus pelagicus, Cyclicargolithus floridanus,
	Pontosphaera multipora, Reticulofenestra bisecta, R. lockeri,
	Sphenolithus moriformis, Zygrablithus bijugatus
Gallician 1 – 1810 m	Coccolithus pelagicus, Sphenolithus moriformis,
	Zygrablithus bijugatus, Reticulofenestra cf. lockeri
Canet 1 – 1100 m	Calcidiscus pataecus, Coccolithus pelagicus, Cyclicargolithus
	abisectus, C. floridanus, Discoaster deflandrei, Helicosphaera
	carteri, Pontosphaera discopora, P. multipora, Reticulofenestra
	bisecta, small reticulofenestrids, Sphenolithus cf. delphix, S.
	moriformis, Zygrhablithus bijugatus
Canet 1 – 1470 m	Calcidiscus pataecus, Chiasmolithus altus, Coccolithus
	pelagicus, Cyclicargolithus abisectus, C. floridanus, Discoaster
	deflandrei, Helicosphaera mediterranea, Pontosphaera
	enormis, P. multipora, Reticulofenestra bisecta, small
	reticulofenestrids, Sphenolithus moriformis, Zygrhablithus
	bijugatus
Canet 1 – 1701 m	Chiasmolithus altus, Coccolithus pelagicus, Cyclicargolithus
	abisectus, C. floridanus, Helicosphaera ethologa,
	Pontosphaera enormis, P. multipora, Zygrhablithus bijugatus,
	Reticulofenestra bisecta, Sphenolithus moriformis,
Maruéjols 1 – 433 m	Coccolithus pelagicus, Cyclicargolithus floridanus,
	Pontosphaera multipora, Reticulofenestra bisecta, R. lockeri,
	Sphenolithus moriformis, Zygrablithus bijugatus
Crossroad	Coccolithus pelagicus, Sphenolithus moriformis,
D716/D981	Helicosphaera sp.

Table S2. Records of calcareous nannofossils.

Pollen and spore floras Site		Can	ot 1		Gallie	sian 1	Gallician 4	Gallician 9	Vauvort 1	Montchamp	Saint-Jean de Maruéjols	Célas Ti	innol	Maruáiole 1	Montoile	Crossroads D716/D981	Sorro do Caunal	Eurot Hill
Taxa Depth (m) or sample name or sample number	1100 m			1701 m	1648 m	1775 m	1580 m	2530 m	3200 m	wontchamp	Pit hill	1	2	798 m	Montens	Crossroads D7 16/D96	3	Euzet Hill
MEGATHERMS (MAT>24°C)* Amanoa (Euphorbiaceae)									1				1			3	2	
Bambusa (Poaceae) Avicennia (Acanthaceae)				1	1					1	3	1	1	1		3		L,
Bombax-type (Malvaceae) Buxus bahamensis-type (Buxaceae)				L.							1		1			1		2
Afzelia-Peltophorum-type (Leguminosae) Canthium-type (Rubiaceae)				1				1		9	2	11				5 1		2
Peltogyne-type (Leguminosae) Croton-Jatropha (Euphorbiaceae)				1						4	2	1				6		1
Euphorbiaceae Leguminosae Caesalpinioideae Malpighiaceae										1		4				1		
Mappianthus (Icacinaceae)				1														3
Mussaenda-type (Rubiaceae) Phyllanthus (Phyllanthaceae)									1				2	1		2	3	2
Rhizophoraceae Rubiaceae										3							1	
Sapindaceae Theaceae									1	2			1		2	2		1
Thunbergia (Acanthaceae) Uncaria-type (Rubiaceae)									6		1		1			1		3
MEGA-MESOTHERMS (20 °C <mat<24 °c)*<br="">cf. Alfaroa (Juglandaceae)</mat<24>									2				1				2	
Alangium (Cornaceae) Arecaceae				4				1	1		1	4	1 2		1			1
Castanopsis-Lithocarpus (Fagaceae) Cissus (Vitaceae)			3									2	5	1	1	1		1
Cyrillaceae-Clethraceae Distylium (Hamamelidaceae)			1	1		1	1	1	2	9	13	2	2	1 1	6	6	3	10
Dodonaea (Sapindaceae) Embolanthera (Hamamelidaceae)									4	4	1	2	1	2	2 33	1		1
Engelhardia (Juglandaceae) Fothergilla (Hamamelidaceae)	3	19	22	18	2	3	1	7	48	9	14	24	13	2	33	8	49	1 54 2
Hamamelis (Hamamelidaceae) Ilex floribunda-type (Aquifoliaceae)												1				1		1
Leea (Vitaceae) Menispermaceae			1							25	3 2	35	23		11	6	1	6
Microtropis (Celastraceae) Myrica (Myricaceae)		2	2			1			1	1			3		1			
Nyssa aquatica-type (Cornaceae) Nyssa cf. sinensis (Cornaceae)	2		E										1					1
cf. Phyllostylon (Ulmaceae) Platycarya (Juglandaceae)			1	2					1	1	2	1	1			1	1	
Rhodoleia (Hamamelidaceae) Rhoiptelea (Juglandaceae)			1						5		16				1			1
Ricinus-type (Euphorbiaceae) Sapotaceae	_1	4	5		2	1			2	1	6	4	7	2	3	11		1
Sciadopitys (Sciadopityaceae) Symplocos (Symplocaceae)	9		1	1					2	2		5	4			5		2
Taxodium-type (Cupressaceae) Cupressaceae papillate pollen grains (former 'Taxodiaceae')	3 44	23	2 43	11 49	7 185	116	140	1 97	2	1 2	51	4	1	46 88	78	39	16 13	
Cathaya (Pinaceae) Podocarpus (Podocarpaceae)	74 24	11	10 30	24 29	55 101	352 134	26 28	22 73	5	6 17	1127 214	5	17	27 64	3 18	3 46	4 89	5 39
MESOTHERMS (14 °C <mat<20 (sapindaceae)<="" acer="" td="" °c)*=""><td><u> </u></td><td>Ť</td><td></td><td>_</td><td></td><td></td><td></td><td></td><td>_</td><td></td><td>16</td><td>_</td><td>Ë</td><td></td><td></td><td>4</td><td>1</td><td>1</td></mat<20>	<u> </u>	Ť		_					_		16	_	Ë			4	1	1
Alnus (Betulaceae)	4	3	1	2							10							
Anacardiaceae Araliaceae Betula (Betulaceae)	1				1	1			1	27		33	37			5	1	
Buxus sempervirens-type (Buxaceae)		1								3	1	2	5					
Carpinus (Betulaceae) Carya Juglandaceae)	10	38	12	6	4	2		6	1			1				2	2	3
Celtis (Cannabaceae) Ericaceae	7	1	1	1	1	1					2		1					
Eucommia (Eucommiaceae) Hedera (Araliaceae)											1						1	2
flex (Aquifoliaceae) Juglans (Juglandaceae) Liquidambar (Altingiaceae)				1														
Lonicera (Caprifoliaceae)			1	3			1	1		3	1	1	3			2	1	
Parrotia (Hamamelidaceae) Populus (Salicaceae)			1	1	1				4	1	6		1			1	1	8
Pterocarya (Juglandaceae) Quercus (Fagaceae)	23	11	22	13	3	2	3	1	6	4	4	3	2	1	2	18	6	1
Salix (Salicaceae) Sambucus (Adoxaceae)																1		1
Taxaceae Ulmus (Ulmaceae)		6		1	2	1		2	4	11	1	2	4		8	1	6	2
Zelkova (Ulmaceae) MESO-MICROTHERMS (12 °C <mat<14 td="" °c)*<=""><td></td><td>19</td><td></td><td></td><td>1</td><td>3</td><td></td><td>1</td><td>2</td><td></td><td>1</td><td></td><td>1</td><td></td><td></td><td></td><td>1</td><td></td></mat<14>		19			1	3		1	2		1		1				1	
Cedrus (Pinaceae) Fagus (Fagaceae) Tsuga (Pinaceae)	12 4		1	2					1					1		4		1
MICROTHERMS (MAT<12 °C)*	6			7	4									3				
Abies (Pinaceae) Pices (Pinaceae)	7 12	7	5	6	2	19 34	1	1 2						1				
TAXA WITHOUT ECOLOGICAL SIGNIFICANCE Pinus diplostellate-type (Pinaceae)	172	9	2	19	11		39	18	3	5	161	1	2	111	2	79		11
Indeterminable pollen grains of Pinaceae Cupressus-Juniperus-type (Cupressaceae)	259 13	109	81 7	135 8	187 17	9427	264	310 9	3 2	4 5	3864 2	5	3	3	37 1	166 1	93	15
Aristolochia (Aristolochiaceae) cf. Cucurbitaceae													1					
Ranunculaceae Rosaceae									4 2				1					4
WATER PLANTS Aglacreidia (fossil taxon)											13						3	
Lemna (Lemnaceae)									4	1							_	
Myriophyllum (Haloragaceae) Nympheaceae				1 4					1	1		1						
Oenotheraceae	1			2						1			1					
Potamogeton (Potamogetonaceae) Restio (Restionaceae) Restionaceae	Ė		2	1	1				3	1						1	2 3	3
SCLEROPHYLLOUS MEDITERRANEAN PLANTS Jasminum (Oleaceae)			Ė										1					Ė
Olea (Oleaceae) Phillyrea (Oleaceae)	1 1	1							10	5	1	5	9			2	16 2	6 2
Quercus ilex-type (Fagaceae) HERBACEOUS PLANTS	Ė	1	4	3													Ĺ	
Amaranthaceae Brassicaceae	4		1	3					1	1	1	1				1	1	1
Campanulaceae	-								3							1		7
Caryophyllaceae Compositae Asteroideae Compositae Cichorioideae	1		4 2									1			1	1		4
Compositae Cicnonolideae Convolvulus (Convolvulaceae) Cyperaceae	1 2		3						2	-1	4	1				13	1 12	1 4
Lyperaceae Lamiaceae Leguminosae Papilionideae	É		j	Ė						3							' <u>'</u>	1
Leguninosae Papilionideae Liliaceae Plantago (Plantaginaceae)	3						1		2	,		4				1		3
Plantago (Plantaginaceae) Plumbaginaceae Poaceae	2 7	1 15	1	3	11	2 9	2	1	19		2	1				2		2
Resedaceae		- 13	Ė	Ľ		,					-	1				-		1 2
Rumex (Polygonaceae) Sanguisorba-type (Rosaceae) Thympleaceae												-				4		1
Thymeleaceae STEPPE PLANTS			Ĺ.,													1		
Artemisia (Compositae) Ephedra (Ephedraceae)		1	8	6	1	2		22			1			4	1			
SUBDESERTIC PLANTS Calligonum (Polygonaceae)			1															
Number of identified pollen grains Unidentified pollen grains Indeterminable pollen grains	721		286					580	165	181 4	5541 1	166 2	167 2	364	212	459	344	238
PTERIDOPHYTE SPORES	10	39	116	37	54	229	303	- 8	20	36	14	43	40 334	2	15	60	17	19
Osmunda (Osmundaceae) Polypodiaceae	1													3			5	
Pteris (Pteridaceae) Unidentified monolete spores	21	4	4	5				1	3	4	2	1	1	5	2	3	128	1 5
Unidentified trilete spores Unidentified spores	12	4	1 4	5	5		3	7 3	1	9 8	1	12	2 2	8 6	3	21	199	4 12
ALGAE Botryococcus										Present	Present	Present	Present		Abundant	Present		Present
REWORKED POLLEN GRAINS AND SPORES Pollen grains	35	4	10	102	ge.	1		205		2	4	18	2	21	4	7	24	Journ
Fern spores	16	2	16 5	3	w			3	1	-	<u> </u>	10		1	<u> </u>	· ·	29	
* MAT (mean annual temperature) with respect to the Nix (1982)'s temper	ure gradi	ent																

Table S3. Detailed content in pollen grains and spores of the analysed samples.

1. Advances with respect to previous palynological records.

Several palynological records were previously performed in the studied region. Most of them are characterised by a listing of the most usually identified plants with a limited or missing counting of pollen grains (Provence: Sittler, 1965; Schuler and Sittler, 1976; Marseilles Basin: Châteauneuf, 1977; Alès Basin: Michoux, 1983). Such studies are particularly damaged by the non-recognition of most of the triporate, tricolpate and tricolporate pollen grains, which often belong to subtropical and tropical plants, i.e. meaningful elements for palaeoenvironmental reconstructions. The use of these poor pollen records is thus very limited for understanding palaeovegetation and palaeoclimate. Even lesser elaborated palynological analyses have been performed on few Camargue wells and do not provide significant information (Jan du Chêne, 1989; Buisson, 1993).

Because of improvements in botanical identification of pollen grains achieved in the seventies on the Neogene pollen floras thanks to the use of the rich ISEM pollen collection in Montpellier (Suc, 1976; Bessedik, 1980, 1981; Suc and Bessedik, 1981), supported by pollen photographs, some progress was introduced in Paleogene microfloras (Célas: Gruas-Cavagnetto, 1973; Provence: Châteauneuf and Nury, 1995), unfortunately still devoid of countings. Accordingly, these pollen records have a limited interest.

Our pollen study confirms the same differences and similarities between the Célas macroflora and microflora than previously indicated by Gruas-Cavagnetto (1973) but emphasizes the important role in the local vegetation made by Hamamelidaceae (*Distylium*, *Embolanthera*, *Rhodoleia*), Sapotaceae, Euphorbiaceae (*Croton*, *Amanoa*, *Canthium*-type) in addition to Araliaceae and Menispermaceae listed in the macroflora. One matter persists due to abundant cones of the fossil genus *Doliostrobus* Marion in the macroflora that Kvacek (2002) ascribed to an extinct family, Doliostrobaceae, expected close of both Araucariaceae and Cupressaceae. Cupressaceae pollen grains (including those of the former Taxodiaceae) are very scarce in the Célas microflora while those of Araucariaceae have not been observed.

2. Dinoflagellate cysts.

Dinoflagellate cysts have been recorded in some samples (Plate 4): Canet 1 (1100 m, 1470 m, 1701 m), Gallician 4 (1580 m), Gallician 9 (2530 m), Crossroads D716/D981, and Serre de Cauvel (samples 1 and particularly 2, the sample in which dinoflagellate cysts are the most abundant). On the whole, dinoflagellate cysts are not well preserved, often broken and thus identifiable with great difficulty. Many reworked dinoflagellate cysts occur. All the individuals have been observed both in transmitted light microscopy (TLM) and under fluorescent UV light microscopy (FLM) in order to detect the reworking (Doláková and Burešová, 2007; Hoyle et al., 2018): the reworked dinoflagellate cysts show a dull brown colour at the difference of the non reworked specimens which show a bright yellow colour as illustrated in Plate 4.

3. Morphological description of some pollen grains.

These descriptions are based on observations at transmitted light microscope (TLM) according to LO-analysis (*Lux* = light vs. *Obscuritas* = darkness) technique and/or scanning electronic microscope (SEM).

Afzelia-Peltophorum-type (Leguminosae) (Plate 1, Figs. 2a-e, 3a-d, 4).

Oblate to equiaxal tricolporate and reticulate pollen grains show large and short non-linear ectoapertures (colpi), endoapertures (pori) being often indistinct. The supratectal reticulum is made of large lumina. Muri are densely micro-echinate. Columellae with large head structurate the tectum, also supported by smaller other ones. This pollen has been recorded in several locations (Canet 1, Gallician 9, Crossroads D716/D981, Euzet Hill; Table S3).

Similar pollen types are found in the tribes Detarioideae (*Afzelia*, *Intsia*) and Caesalpinioideae (*Peltophorum*, *Delonix*) (Plate 1, Figs. 1a-e; Banks and Lewis, 2018) but none of the observed species from the ISEM Collection shows small spines.

Peltogyne-type (Leguminosae; Plate 3, Figs.6a-d, 7).

Oblate tricolporate and clavate pollen with thick costae along the ectoapertures (colpi) and a distinct thin annulus bordering the endoapertures (pori). Large and smaller clavae constitute the ornamentation of the pollen. This pollen has been recorded in the Euzet Hill location.

This pollen morphology is similar to that of the modern species *Peltogyne confertiflora* (Hayne) Benth. (Detarioideae) (Graham and Barker, 1981: plate 12, fig. E).

Croton-Jatropha (Euphorbiaceae; Plate 2, Figs. 3, 5a-b).

Inaperturate spheroidal pollen grain with a coarsely reticulate ornamentation constituted by more or less dense clavae, which have an almost triangular shape that provides the so-called *Croton*-pattern. This pollen has been recorded in several locations (Canet 1, Montchamp, Saint-Jean de Maruéjols Pit hill, Célas Tunnel, Crossroads D716/D981, Euzet Hill; Table S3). Such very peculiar pollen types are characteristic of *Croton* (Plate 2, Figs. 4a-c) and *Jatropha* genera (Carreira et al., 1996; Ribes de Lima, 2007; Ribeiro de Souza et al., 2020).

Amanoa (Phyllanthaceae; Plate 1, Figs. 6a-c).

Tricolporate oblate pollen showing a large semi-tectate curvilinear reticulum (a thin membrane covers the lumina), with long colpi (ectoapertures) resulting in a small apocolpium, bordered by costae. Endoapertures are rectangular. This pollen has been recorded in several locations (Vauvert 1, Célas Tunnel, Crossroads D716/D981, Serre de Cauvel; Table S3).

This pollen morphology is characteristic of the genus *Amanoa* (Plate 1, Figs. 5a-b; Punt, 1962; Köhler, 1965).

Dodonaea (Sapindaceae; Plate 2; Figs. 6a-b).

Equiaxal tricolporate pollen, subtriangular in polar view, with prominent apertural areas characterized by thickening endexine, especially around the rectangular endoapertures. The ornamentation can be psilate, rugulate or slightly reticulate. This pollen has been recorded at Célas Tunnel and Crossroads D716/D981 locations (Table S3).

Because of its distinctive shape, this pollen corresponds in part to the fossil 'genus' *Pentapollenites* conceived by Krutzsch (1962), manifestly illustrated at the scanning electronic microscope by Châteauneuf (1980: plate 17, fig. 7), which now receives a robust botanical ascription as *Dodonaea* (Plate 2, Figs. 7a-c; Merville, 1965), a primitive genus within the Sapindaceae (Muller and Leenhouts, 1976). Henceforth, the name *Pentapollenites* must be rejected as it probably also includes other botanical genera.

Alangium (Cornaceae; Plate 1, Figs. 7, 8).

Prolate tricolporate and reticulate pollen, elliptic in equatorial view, with long ectoapertures (colpi) edged by thick costae. Lalongate endoapertures (pori) are surrounded by a thin annulus. The supratectate reticulum is somewhat lengthened along the pole axis. The tectum is thick and clearly separated from the endexine. This pollen has been recorded at Vauvert 1, Célas Tunnel, Saint-Jean de Maruéjols Pit hill and Monteils locations (Table S3).

Such a pollen is characteristic of several species of the *Alangium* genus (Plate 1, Fig. 9; Reitsma, 1970).

Plate 1. Photographs at TLM and SEM of some fossil and modern pollen grains (shots by J.-P. Suc). Modern pollen grains originate from the ISEM - Montpellier Collection.

Scale bar = $10 \mu m$.

Figs 1a-e. *Afzelia cochinchinensis* (Pierre)J.Léonard = *A. xylocarpa* (Kurz)Craib (Leguminosae) – ISEM Collection number: 9427 (plant specimen collected in Vietnam); equatorial view aperture facing at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the head of columellae; c, focus on the foot of columellae; d, optical section.

Figs. 2a-e. Afzelia-Peltophorum-type (Leguminosae) – Euzet Hill; equatorial view (intercolpium) at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the head of columellae; c, focus on the foot of columellae; d, aperture; e, optical section.

Figs. 3a-d. *Afzelia-Peltophorum*-type (Leguminosae) – Crossroad D716/D981; equatorial view (intercolpium) at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the head of columellae; c, focus on the foot of columellae; d, optical section.

Fig. 4. Afzelia-Peltophorum-type (Leguminosae) – Euzet Hill; equatorial view aperture facing at SEM.

Figs. 5a-b. *Amanoa caribaea* Krug & Urb. (Phyllanthaceae) – ISEM Collection number: 41784 (plant specimen collected in Cameroon); equatorial views aperture facing (a) and polar view (b) at TLM.

LO-analysis: a, reticulate ornamentation and apertures; b, optical section.

Figs. 6a-c. *Amanoa* (Phyllanthaceae) – Crossroad D716/D981; equatorial view aperture facing (a-b) and polar view (c) at TLM.

LO-analysis: a, reticulate ornamentation and aperture; b, optical section.

Fig. 7. Alangium (Cornaceae) – Célas Tunnel 1; equatorial view (intercolpium) at TLM.

Fig. 8. Alangium (Cornaceae) - Saint-Jean de Maruéjols Pit hill; equatorial view aperture facing at TLM.

Fig. 9. *Alangium barbatum* (R.Br. ex C.B.Clarke) Baill. ex Kuntze (Cornaceae) – ISEM Collection number: 17360 (plant specimen collected in Burma); equatorial views aperture facing and optical section at TLM.

Figs. 10a-b. Bombax-type (Malvaceae) – Crossroad D716/D981; polar view at TLM.

LO-analysis: a, reticulate ornamentation; b, optical section.

Figs. 11a-b. Avicennia (Acanthaceae) - Montchamp; equatorial view aperture facing at TLM.

LO-analysis: a, reticulate ornamentation; b, optical section.

Figs. 12a-c. Avicennia (Acanthaceae) – Saint-Jean de Maruéjols Pit hill; equatorial view (intercolpium) at TLM.

LO-analysis: a, reticulate ornamentation; b, focus at base of columellar, colpi; c, optical section.

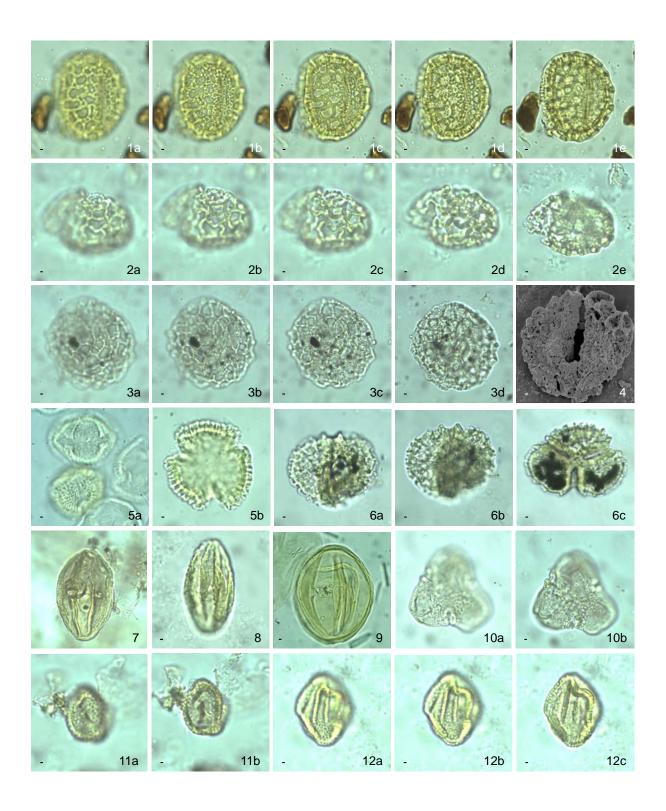


Plate 2. Photographs at TLM and SEM of some fossil and modern pollen grains (shots by J.-P. Suc). Modern pollen grains originate from the ISEM - Montpellier Collection or from the GeoBioStratData - Rillieux la Pape Collection.

Scale bar = $10 \, \mu m$.

Figs 1a-b. Araliaceae – Célas Tunnel 2.; equatorial view (intercolpium) at TLM.

LO-analysis: a, reticulate ornamentation and apertures; b, optical section.

Figs. 2a-b. Araliaceae – Montchamp; equatorial view aperture facing at TLM.

LO-analysis: a, reticulate ornamentation and apertures; b, apertures and optical section.

Fig. 3. Croton (Euphorbiaceae) – Euzet Hill; view at SEM.

Figs. 4a-c. *Croton scabiosus* Bedd. (Euphorbiaceae) – ISEM Collection number: 15339 (plant specimen collected in India); view at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the foot of columellae; c, optical section.

Figs. 5a-b. Croton (Euphorbiaceae) – Saint-Jean de Maruéjols Pit hill; view at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the foot of columellae and optical section.

Figs. 6a-b. Dodonaea (Sapindaceae) - Crossroad D716/D981; equatorial view aperture facing at TLM.

LO-analysis: a, aperture; b, aperture and optical section.

Figs. 7a-c. *Dodonaea triquetra* J.C.We,dl. (Sapindaceae) – ISEM Collection number: 36963 (plant specimen collected in Australia); equatorial view aperture facing at TLM.

LO-analysis: a, psilate ornamentation and apertures; b, aperture; c, aperture and optical section.

Figs. 8-c. *Embolanthera glabrescens* H.L.Li (Hamamelidaceae) – ISEM Collection number: 14245 (plant specimen collected in Vietnam); polar view at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the foot of columellae; c, optical section.

Figs. 9a-c. Embolanthera (Hamamelidaceae) – Vauvert 1 3200 m; polar view at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the foot of columellae; c, optical section.

Figs. 10a-c. *Fothergilla gardenii* L. (Hamamelidaceae) – GeoBioStratData Collection number: 839 (plant specimen collected in North America); polar view at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the foot of columellae; c, optical section.

Figs. 11a-c. Fothergilla (Hamamelidaceae) – Euzet Hill; polar view at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the foot of columellae; c, optical section.

Figs. 12a-b. Mappianthus (Icacinaceae) – Euzet Hill; equatorial view at TLM.

LO-analysis: a, echinulate ornamentation and aperture; b, optical section.

Fig. 13. Leea (Vitaceae) – Saint-Jean de Maruéjols Pit hill; equatorial view at TLM.

LO-analysis: reticulate ornamentation and aperture.

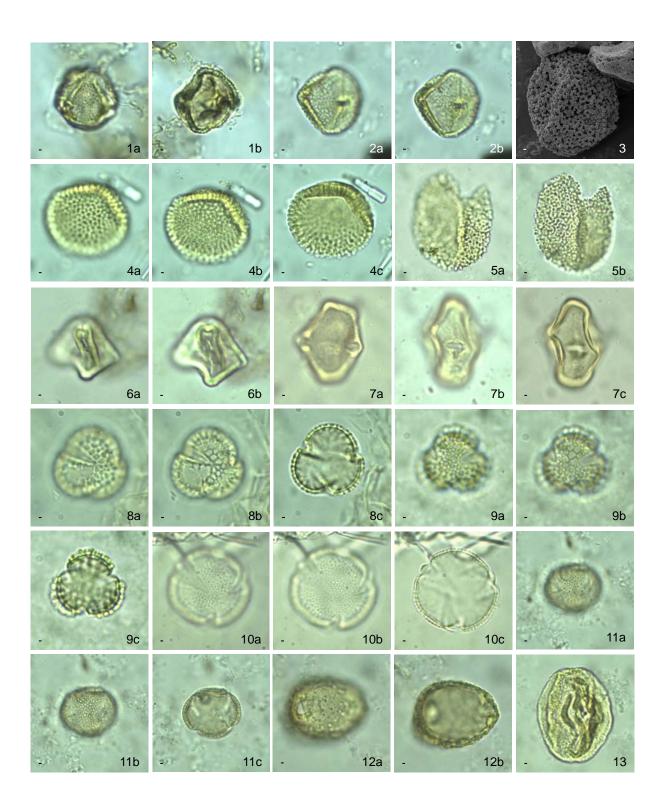


Plate 3. Photographs at TLM and SEM of some fossil and modern pollen grains (shots by J.-P. Suc).

Modern pollen grains originate from the ISEM - Montpellier Collection.

Scale bar = $10 \, \mu m$.

Figs. 1a-e. *Distylium* (Hamamelidaceae) – Euzet Hill; polar view at TLM.

LO-analysis: a, reticulate ornamentation; b, apertures and optical section.

Figs. 2a-c. Menispermaceae – Euzet Hill; equatorial view aperture facing at TLM.

LO-analysis: a, reticulate ornamentation and aperture; b, focus on the base of columellae; c, optical section.

Figs; 3a-c. Olea (Oleaceae) – Euzet Hill; equatorial view (intercolpium) at TLM.

LO-analysis: a, reticulate ornamentation; b, focus on the base of columellae; c, optical section.

Fig. 4. *Mussaenda laxa* (Hook.f.)Hutch. ex Gamble (Rubiaceae) – ISEM Collection number: 9954 (plant specimen collected in India); polar view, optical section at TLM.

Fig. 5. Mussaenda-type (Rubiaceae) – Serre de cauvel 3; polar view, optical section at TLM.

Figs. 6a-d. *Peltogyne*-type (Leguminosae) – Euzet Hill; polar view at TLM.

LO-analysis: a, clavate ornamentation; b, focus on the base of clavae; c, apertures; d, optical section.

Fig. 7. *Peltogyne*-type (Leguminosae) – Euzet Hill; view with aperture at SEM.

Figs. 8a-c. *Phyllanthus simplex* Retz. = *P. virgatus* G.Forst. (Phyllanthaceae) – ISEM Collection number: 10100 (plant specimen collected in India); polar view at TLM.

LO-analysis: a, reticulate ornamentation and apertures; b, focus on the base of columellae and apertures; c, optical section.

Figs. 9a-c. *Phyllanthus* (Phyllanthaceae) – Euzet Hill; polar view at TLM.

LO-analysis: a, reticulate ornamentation and apertures; b, focus on the base of columellae and apertures; c, optical section.

- Fig. 10. Phyllanthus (Phyllanthaceae) Euzet Hill; view at SEM showing the reticulum and apertures.
- Fig. 11. Rhizophoraceae Montchamp; equatorial view at TLM showing the apertures.
- Fig. 12. Rhoiptelea (Juglandaceae) Canet 1 1470 m; polar view, optical section at TLM.
- Fig. 13. Symplocos (Symplocaceae) Euzet Hill; polae view showing apertures at TLM.
- Fig. 14. Cedrus (Pinaceae) Crossroad D716/D981; profile view at TLM.
- Fig. 15. Podocarpus (Podocarpaceae) Célas Tunnel 2; proximal view at TLM.
- Fig. 16. Podocarpus (Podocarpaceae) Crossroad D716/D981; distal view at TLM.

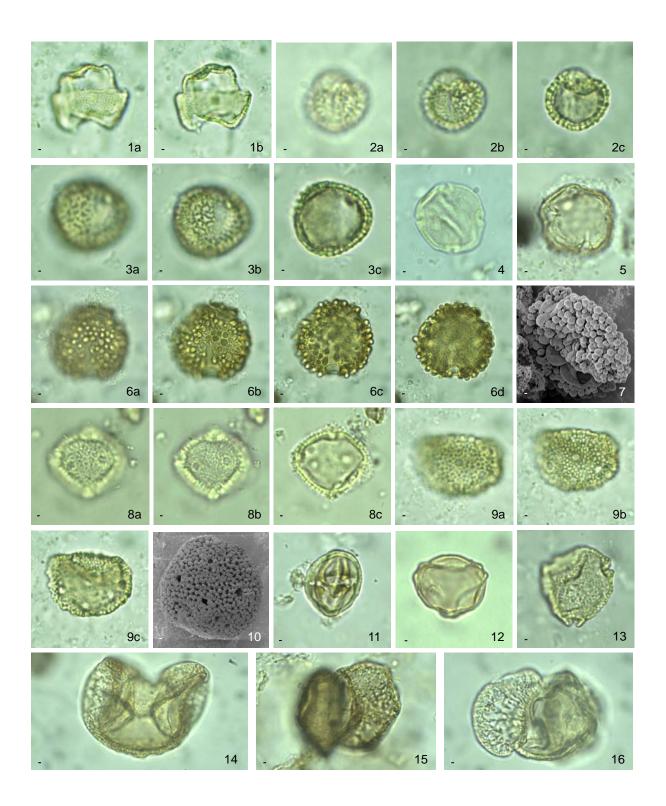


Plate 4. Photographs at TLM and FLM of some fossil dinoflagellate cysts (shots by S.-M. Popescu).

Scale bar = $10 \mu m$.

Figs. 1a-c. Deflandrea phosphoritica – Gallician 9 (2530 m): a-b, TLM; c, FLM.

Figs. 2a-c. Spiniferites sp. – Gallician 9 (2530 m): a-b, TLM; c, FLM.

 $Figs.\ 3a-c.\ \textit{Nematosphaeropsis}\ sp.-Crossroad\ D716/D981:\ a-b,\ TLM;\ c,\ FLM.$

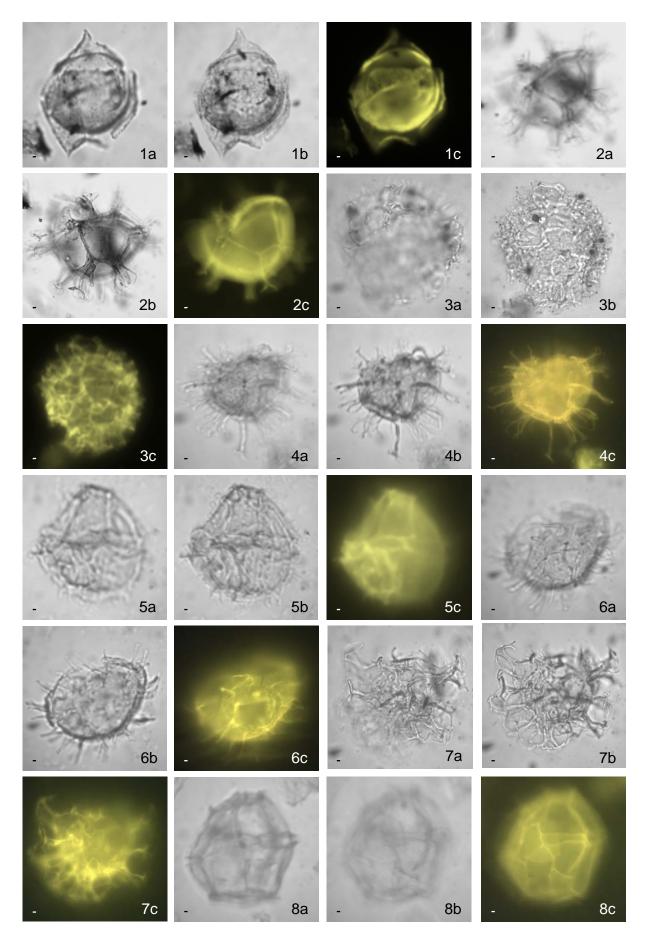
Figs. 4a-c. Cleitosphaeridium sp. – Serre de Cauvel (sample 2): a-b, TLM; c, FLM.

Figs. 5a-c. Impagidinium sp. – Serre de Cauvel (sample 2): a-b, TLM; c, FLM.

Figs. 6a-c. Lingulopdinium machaerophorum – Serre de Cauvel (sample 2): a-b, TLM; c, FLM.

Figs. 7a-c. Glaphyrocysta sp. – Canet 1 (1100 m): a-b, TLM; c, FLM.

Figs. 8a-c. Impagidinium patulum – Canet 1 (1100 m): a-b, TLM; c, FLM.



4. Climatostratigraphy vs. palynostratigraphy.

Two obvious items imply to use pollen data as a climatostratigraphic tool (numerical variability) in contrary to a classical biostratigraphic tool (first occurrence and last occurrence datums):

- plants evolve along geological times but evolution only affects the species level during the Cenozoic period, usually unattainable through the pollen morphology;
- plants shift in latitude and altitude along geological times with respect to changes in temperature and precipitation.

As a consequence, palynostratigraphy based on occurrences of supposed species is to be considered as an old-fashioned approach and must be definitely abandoned. One of the most critical examples of such an obsolete use is provided by the use of the pseudo-species *Boelhensipollis hohli*, which has been proposed as a marker of the Oligocene series (Sittler and Schuler, 1976) and hence abundantly indeed excessively exploited (e.g.: Châteauneuf, 1980; Olivier-Pierre, 1980; Schuler, 1988; Châteauneuf and Nury, 1995; Cavagnetto, 2002; Châteauneuf et al., 2019).

The problem resulting from this deficient concept shows multiple aspects:

- quality of the published photographs is often insufficient to convince that an unique taxon corresponds to the so-called *Boelhensipollis hohli* pollen;
- Sittler and Schuler (1976) suppose that two pollen-types are included within their *Boelhensipollis hohli* that makes more inappropriate the use of a species designation;
- these authors describe the *Boelhensipollis hohli* pollen-type as heteropolar, i.e. non-joined colpi (strictly tricolporate pattern) on one side of the polar view and joined colpi (syncolporate pattern) on the other side, and they propose two possible botanical ascriptions: *Elaeagnus* (Elaeagnaceae) and *Serjania* (Sapindaceae); *Elaeagnus* modern species do not show heteropolar pollen while some *Serjania* modern species show heteropolar pollen (Muller and Leenhouts, 1976);
- in addition, it is difficult to conceive the sudden appearance of the temperate genus *Elaeagnus* at the earliest Oligocene, while it has been already recorded in Northern Canada during the Early Eocene Climatic Optimum (Salpin et al., 2019);

it is also unbelievable that *Serjania*, a tropical genus, appeared simultaneously with a significant climate cooling, while several pollen grains belonging to Sapindaceae have been recorded in the Priabonian samples (Table S3) and are present in Eocene sediments from Southwestern France (Fauquette et al., in progress).

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GEOCHEMICAL MODELLING

	Late Oligocene seawater end-member	Riverine end-member
⁸⁷ Sr/ ⁸⁶ Sr	0.70825	0.7077
δ ³⁴ S VCDT	20	11
[Sr]	15	0.1
[SO ₄ ²⁻]	2800	70

Table S4. Sulphur and Strontium isotope composition and concentrations for the late Oligocene seawaterand riverine end-members (best-case scenario).