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ORGANIC CHARACTERIZATION OF THE "TÉGULINES CLAY FORMATION", EAST PARIS-BASIN, FRANCE: A BULK AND MOLECULAR INVESTIGATION

Le Meur Mathieu^{a,b,c}, Boussafir Mohammed^{a,b,c}, Le Milbeau Claude^{a,b,c}, Debure Mathieu^d, Claret Francis^d, Robinet Jean-Charles^e & Lerouge Catherine^d

(a) Université d'Orléans, ISTO, UMR 7327, 45071 Orléans, France; (b) CNRS, ISTO, UMR 7327, 45071 Orléans, France; (c) BRGM, ISTO, UMR 7327, BP 36009, 45060 Orléans, France; (d) BRGM, BP36009, 45060 Orléans Cedex 2, France; (e) ANDRA 1-7 rue Jean Monnet 92298 Chatenay Malabry, France

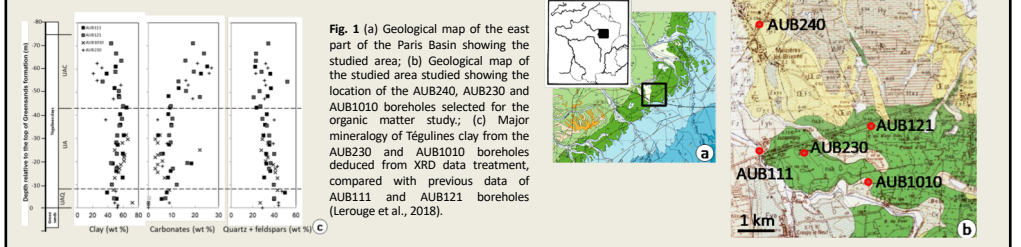


Introduction

The Tégulines Clay formation (Paris Basin, France) is currently under investigation as a potential host for sub-surface geological repository of low-level long-lived radioactive waste. Lerouge et al. (2018, Doi.org/10.1155/2018/1606753) characterized the chemical, mineral, petrological, and textural properties of this rock highlighting a potential reactivity of the organic matter (OM) due to weathering processes. In order to get a global overview of the potential interaction of the Tégulines Clays with radionuclides, a OM study is needed. Our objective was to fully characterize the quantity and quality of the OM of this marine clay formation. Indeed, the refractory or reactive character of OM is an indispensable data for evaluating the overall geochemical reactivity of the rock formation and its long-term evolution.

Setting, material and methods

Several cores and trenches were performed between October and December 2017 in the "Brienne-le-Chateau area" (Aube department, France). Several techniques, ranging from bulk to molecular scales were used in order to get a global characterization of the OM fraction. Rock-Eval (RE) analysis permit us to get several parameters such as the Total Organic Carbon (TOC) content, Hydrogen Index (HI) or the Oxygen Index (OI). The quantitative organic petrography (palynofacies) consists on the microscopic observation of isolated OM, permitting the identification and the quantification of the organic compounds present in a constant amount of sediments (1 gram) after the elimination of carbonate and silicate by hydrochloric and hydrofluoric treatments. Finally, in order to get information at molecular scale, the labile fraction (lipids) of sediments was extracted from dried sediments, fractionated into neutral, acidic and polar compounds, derivatized using BSTFA and analyzed by GCMS.



Results

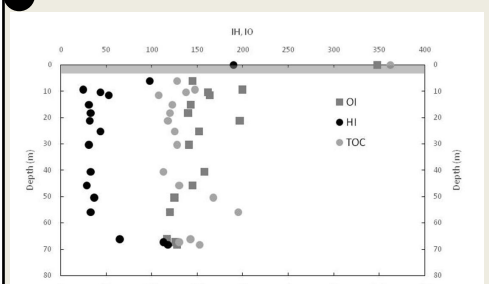
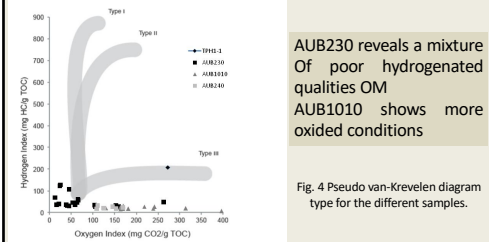


Fig. 3 Hydrogen Index (HI, mg HC.g⁻¹TOC), Oxygen Index (OI, mg CO₂.g⁻¹TOC, values +80 for more clarity) and Total Organic Carbon (TOC, %) in relation with depth for the AUB230 borehole (The sample TPH 1-1 0-30 was also incorporated). The grey part correspond to the superficial formation (TPH 1-1)

- From a general overview, the TOC values show very poor organic content. The TPH 1-1 trench shows extremely low TOC values except for the surface sample (0 – 30 cm). The rest of the trench does not contain any analyzable OM. The TPH 1-1 sample (0-30) is characterized by high HI and OI values. These values are typical of soils and reveals immature OM.

- RE parameters measured for the AUB230 core allows discriminating three main parts corresponding to the top (6 – 9 m), the center (10 – 45 m) and the bottom of the borehole (55 – 68 m). At the top of the borehole, (6 – 9 m), HI decreases (in relation to the surface) and OI increases at 9 m. This opposite behavior suggests more oxidated OM.



Extraction yield	Major alkanes ^a	total (µg/g sed) ^b	odd/ev en ^c	CPI ^d
TPH1-1 0-30cm	n-C29, n-C31, n-C33	5.08	2.97	4.95
AUB230 9.36m	n-C29, n-C31, n-C32	9.14	1.26	1.08
AUB230 21.30m	n-C29, n-C31, n-C33	0.95	1.06	0.97
AUB230 25.32m	n-C22, n-C29, n-C31	0.61	1.37	1.19
AUB230 45.84m	n-C29, n-C31, n-C33	0.56	1.34	1.15
AUB230 66.35m	n-C29, n-C31, n-C33	0.47	0.97	1.57
AUB230 67.45m	n-C25, n-C30, n-C33			

- The AUB230 borehole is characterized by low content in labile compounds. Between 6 m to 21 m, the yields decrease suggesting more oxidated conditions. At 25 m, the yield increases suggesting more preserved conditions at this depth. The labile OM yields at the end of the borehole vary between 0.7 and 1 mg.g⁻¹ showing more preserved conditions.

- The CPI is high for the trench sample and decrease after to values around 1. The CPI values are high at the top, suggesting more preserved OM more degraded conditions between 6 and 21 m and again, more preserved conditions at the end of the borehole.

- The Ref-FAs, are more present at the end of the borehole revealing more preserved conditions. In addition, a-C15:0, i-C15:0 are present in the samples at 6 and 9 m, revealing more bacterial contribution.

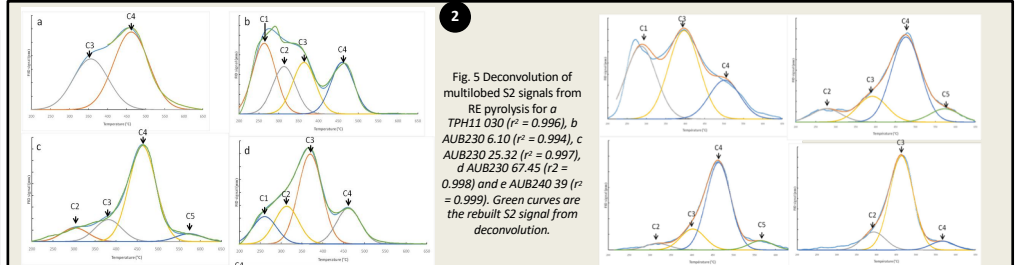


Fig. 5 Deconvolution of multilobed S2 signals from RE pyrolysis for a TPH11 030 (r² = 0.996), b AUB230 6.10 (r² = 0.994), c AUB230 25.32 (r² = 0.997), d AUB230 67.45 (r² = 0.998) and e AUB240 39 (r² = 0.999). Green curves are the rebuilt S2 signal from deconvolution.

- The AUB1010 borehole shows a S2 deconvolution results comparable to the AUB230 borehole. The top of the profile shows more immature OM (contribution of C1).

- The 0-30 cm sample (TPH1-1) is typical of the thermal breakdown of biological constituents such as polysaccharides and lignin and is typical of a soil sample.

- For all samples of the AUB230 borehole (except at 6.10, 67.45 and 68.45 m deep) the most represented cluster is C4 which represent the more stable organic compounds. The presence of a unimodal Gaussian S2 curve with higher contribution of C4 cluster suggests the presence of more stable OM.

- The deconvolution of the S2 curve shows a multilobed shape and suggests the presence of different kinds of OM or the presence of OM with different states of degradation. Between 10 and 55 m, the S2 curve shape shows more contribution of the C4 cluster and suggests more homogenous conditions with an intermediate oxidation state.

- All the S2 peak deconvolution for the AUB240 borehole samples are similar. The C1 cluster is also represented revealing more preserved conditions.

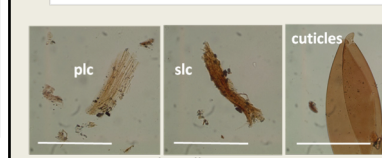
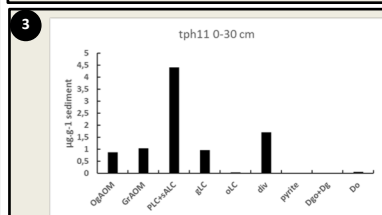


Fig. 7 Distribution of the different particles in TPH1-1 0-30 cm sample and images of the particles encountered, scale: 100 µm

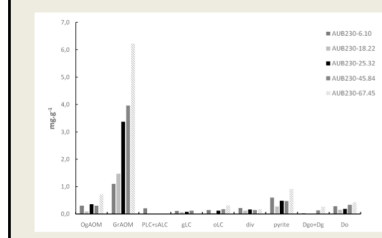


Fig. 8 Distribution of the different particles in the AUB230 borehole (6.10, 18.22, 25.32, 45.84, 67.45 m) and images of the particles encountered, scale 100 µm

- The TPH1-1 sample reveals a predominance of preserved ligno-cellulosic fragments, slightly degraded ligno-cellulosic fragments and divers particles.

- In the borehole AUB230, at 6.10 m, the palynofacies is dominated by grAOM. There is the presence of pyrite with small particles (between 10 and 20 µm). In addition, the div category decreases. The palynofacies at this depth is characterized by small particles (around 1 µm) revealing more oxidative conditions.

- At 25 m, the proportion of grAOM and pyrite increase. At the end of the borehole, (55-68m), the proportion of grAOM is the highest and the grAOM proportion is also high with bigger size and pyrite inclusion showing more preserved conditions. The palynofacies shows higher contribution of grAOM and ogAOM with pyrite inclusion. This feature also shows more preserved conditions.

- The AUB240 borehole showed homogeneous palynofacies content and is characterized by the presence in majority of grAOM and ogAOM. There is also the presence of pyrite in inclusion in the ogAOM suggesting more preserved conditions.

- The AUB1010 borehole palynofacies shows small particles (1 µm) for all samples suggesting more oxidative conditions

Conclusion: This study permitted to characterize the OM oxidation profile of the Tégulines Clay formation and to compare it with previous mineralogical, geological and petrophysical data. The organic investigation showed, for the Trench TPH1-1 oxidative conditions except for the surface sample. The AUB230 and AUB1010 borehole showed a gradual oxidation profile along the core and for the AUB230 core some preserved areas. The AUB240 borehole revealed more preserved conditions. All the samples were characterized by low content in OM. This low content was associated with more refractory conditions, supposing a limited influence of the OM on the transfer and reactivity of radionuclides.

For further information, here is the flashcode of my researchgate webpage. E.mail address :m.lemeur@hotmail.fr