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Weathering profile in a near-surface aquitard : example of the teguline Clay, East Paris Basin, France

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Near-surface aquitards are targeted for the containment of low-level radioactive waste (LLW) in near-surface environments because they could act as natural low-permeability protective barriers for underlying groundwater resources. Few detailed mineralogical and geochemical studies have been dedicated to surficial clay formations (Hendry and Wassenaar, 2000), compared to the abundance of data made available for reduced clay formations foreseen for deep nuclear repository systems. Their study is however of primary interest for the near-surface waste storage. The objectives of this study were (1) to describe the distribution of major ions in pore waters of a 80 m-thick clay-rich aquitard near the surface, and (2) to identify geochemical processes controlling the distribution of these ions. These objectives were completed by measurements of several parameters which are currently used to model pore water chemistry in deep reduced aquitards: (1) mineralogy and diagenetic sequence, (2) water content and porosity, (3) cation exchange capacity and cation distribution on the clay exchanger as well as the determination of anion concentrations (Cl and SO<sub>4</sub>) through milliQ leaching, (4) partial pressure of CO<sub>2</sub>, O<sub>2</sub>, and N<sub>2</sub>.

The investigated area was located in the eastern part of the Paris Basin. The Aptian Gault clay outcrops on a 8-10 km large and 80-km long band of terranes through the Aube department. Two drilling campaigns were conducted by Andra in the area of Brienne Le Château through surficial formation and the Gault Clay formation down to Greensands. In the studied area, surficial formations are not deeper than 6 m, and the Gault Clay is only represented by the teguline Clay. The thickness of the clay formation crosscut through seven boreholes ranges from almost 10 m in the south-east to nearly 65-75 m in the north-west.

The mineralogy consists of a dominant clay fraction (illite, illite-smectite mixed layers, chlorite, kaolinite, glauconite) associated with a quartz - K feldspar silty fraction and a carbonate fraction (calcite, dolomite, siderite). Accessory minerals are represented by nodules of phosphates, traces of celestine and framboidal pyrite. Diagenetic minerals indicate an evolution of marine clay sediments under reducing conditions at the limit between the suboxic zone (siderite) and the bacterial sulphate reduction zone (pyrite). Weathering is marked by an abrupt color change from brownish on the first meters to greenish in the deeper part of the formation (Fe<sup>2+</sup>/Fe<sup>3+</sup> transition), a partial dissolution of K-feldspar and calcite, a destabilization of ankerite, and an oxidation of pyrite in gypsum, iron sulphates and iron hydroxides. Some vertical fractures and some plans parallel to the bedding that are coated by iron hydroxides were observed down to ~9 meter depth.

The bulk rock chemistry (major and minor elements) and the cation exchange capacity were slightly modified by weathering. The proportions of exchangeable cations were stable with depth the exception of an increase of Mg concentrations at expense of Ca towards the surface. Chloride concentrations were low, varying a little with depth, but varying as a function of the location of the boreholes. These lateral variations were not observed for sulphate that displayed in turn increasing concentrations from ~ 7 mmole/L pore water to up to 38 mmole/L pore water toward the surface (<20 m) when gypsum was not observed.

Gases (CO<sub>2</sub>, O<sub>2</sub>, N<sub>2</sub>, alkanes) were measured by degassing core samples conditioned in cells under He atmosphere. The cores were regularly sampled during the drilling of the AUB1010 borehole down to a depth of 40 m. Moreover few samples taken at various depths from other boreholes were also monitored. The data profiles obtained on samples from different borehole were consistent. CO<sub>2</sub> partial pressure (P<sub>CO2</sub>) reached a steady state after ~2 months and ranged values between 4 and 50 mbar. The highest P<sub>CO2</sub> (20-50 mbar with a mean value of 31 ± 11 mbar) were attained for shallow depth lower than 16 m, whereas average P<sub>CO2</sub> values of 9 ± 3 mbar were established at depths higher than 16 m. Near surface (~2 m) the P<sub>CO2</sub> was once again equal to 10 mbar. It is noteworthy that when the gas cell had a leak, oxygen/nitrogen ratio was close to the atmosphere for samples near surface whereas oxygen was systematically consumed by cores sampled below ~4 m and measured P<sub>CO2</sub> was higher than normal value. After three successive gas monitorings with reconditioning of the cell conducted on core samples of the AUB1010 borehole, P<sub>CO2</sub> measured in all the samples became homogeneous with an average P<sub>CO2</sub> value of 9 ± 3 mbar (figure 1).

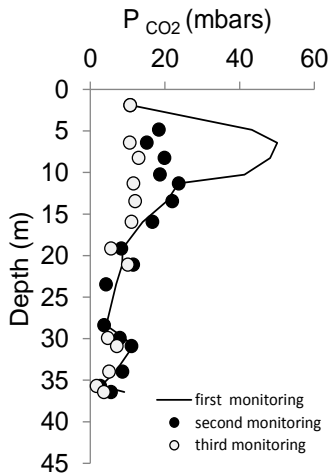


Figure 1:  $P_{CO_2}$  in function of depth during the three successive monitorings

The major identified processes of weathering were thus associated to 1) pyrite oxidation and 2) ankerite destabilization. Pyrite oxidation released sulfates and iron, inducing formation of iron hydroxides and gypsum at the expense of Ca from the clay exchanger. Iron sulphates formation occurred more rarely. The pH decrease displaced the carbonate system by  $P_{CO_2}$  increasing. The return of  $P_{CO_2}$  to the reference value ( $9 \pm 3$  mbars) encountered at the bottom of the formation after three successive monitorings indicated that oxygen disappearance from the clayrock brought back the system (clayrock and carbonates) to a gas/rock/pore water equilibrium. Consequently high  $P_{CO_2}$  values can be considered as the record of the clayrock reactivity to oxygen diffusing from the surface.

Hendry, M.J. and Wassenaar, L.I. (2000). Controls on the distribution of major ions in pore waters of a thick surficial aquitard. *Water Resources Research* 36, 503–513.