Miocene cryptokarsts of Entre-Sambre-et-Meuse and Condroz plateaus.

Paleoenvironment, evolution and weathering processes

Christian DUPUIS (1)
Dominique NICAISE (2)
Thierry DE PUTTER (1)
Alain PERRUCHOT (3)
Muriel DEMARET (4)
Emile ROCHE (4)

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Introduction

The Dinantian limestones of Entre-Sambre-et-Meuse (ESEM) and Condroz plateaus are scattered with numerous (>350 recensed) large karstic cavities filled with sands, clays, gravels and lignite of Eo-Oligocene and Mio-Pliocene ages. The extraction of raw materials in quarries or in underground mines revealed uncommon dimensions for such karstic cavities, up to 500 m in diameter, around 70-150 m in depth. Since the 19th century geologists have drawn attention to these “vallées d’effondrement” (Van Den Broeck et Rutot, 1888), for example Bayet, 1896; Briart, 1888; d’Halloy, 1841; Lohest, 1887, 1896.

During the first half of the 20th century, when the extraction was still very active, extensive studies were undertaken by Calembert who collected crucial data of the structure of these paleokarsts (1942-3; 1944-5; 1947; 1959). Soyer (1972, 1978) focused on the sedimentology of the Tertiary sands of the ESEM and investigated the palynological content of the organic sediments with Schuler and Sittler. Deduced paleoenvironmental reconstructions have significantly contributed to the age determination of the karst infilling and produced chronological data that are always useful and have updated the prior documentation due to Gilkinet (1922) and Stockmans et Williere’s studies (1934).

The discovery of halloysite occurrences in the ESEM (Ertus *et al*., 1989) significantly after the one evidenced by Buurman and Van der Plas (1968) in the Condroz encouraged to promote approaches focusing on paleoweathering mineralogical and geochemical processes related with the karstic development (Ertus, 1990; Brouard,1992; Dupuis and Ertus,1993,1994; Lemy,1996; Perruchot *et al*., 1997; Nicaise, 1998; De Putter *et al*.1996; De Putter *et al*., 2002). Complementary paleoenvironmental and chronological data were obtained through carpological and palynological studies (Fairon-Demaret, 1992; 1994; Russo Ermolli, 1991). Currently these karsts are used as case studies to model surface geochemical processes related with the migration of cold acid fluids and correlative sequences of mineral neogenesis.

Trapped sediments, paleoenvironments and ages

The first unit of the paleokarst infilling is the sandy Onhaye Formation, the variable thickness of which (~0-20 m) is related to erosion and/or lateral variations. Grain size distribution, sporadically preserved glauconite and *Ophiomorpha* burrows (Gulinck, 1963, 1966; Ertus, 1990) testify to its marine origin. Clays layers that are known place to place at the base of the unit (Nicaise, 1998), have yielded palynomorphs and dinokysts of lowermost Oligocene in the Oret sand pit (Schuler and Sittler in Soyer, 1978). Re-examination of the palynological data confirms this stratigraphic position near the Eocene-Oligocene limit, but does not fully clarifies the correlation of the basal sand with the two potential equivalent beds in northern Belgium: the Watervliet Clay and the transgressive Grimmertingen Sands (Steurbaut, 1992; De Coninck, 1995, 1996).

Above, are piled up 60 to 100 m of variable types of sediments that reveal diverse paleoenvironments: sands and gravels of river channels, clays and silty clays of flood plains, “varved” clays and silts of ponds and lakes, organic clays of rooted vegetation soils, charcoal bearing-silt, probably related to wild fire, deposited in shallow ponds or at least at change of incline (Ertus, 1990; Brouard, 1992;

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(1) Géologie Fondamentale et Appliquée, Faculté Polytechnique, rue de Houdain, 9. B-7000 Mons, Belgium. christian.dupuis@fpms.ac.be.
(2) CSTC-WTCB Research Institut (BBRI), Avenue Holoffe, 21. B-1342 Limelette, Belgium.
(3) Géochimie des roches sédimentaires, Université Paris-sud, Bât.504. F-91405 Orsay Cedex, France.
(4) Paléobotanique et palynologie, Université de Liège, Sart Tilman, Allée du 6 Août, B18. B-4000 Liège, Belgium.

Meyer and Wells, 1997). Lignites deposition needs a special mention. Most of the karstic traps contains accumulations of plant organic matter rich in trunk segments and pieces of *Taxodium* randomly mixed with an amorphous lignitic matrix. Such a potential coal, sporadically prospected and sometimes dug, is locally known as “Machuria” (Calembert, 1947; Nicaise, 1998).

Other fossils lacking, plant remains are very useful to decipher chronological and paleoenvironmental informations. Past paleoenvironmental-chronological attributions used leaves and wood pieces, but currently seeds and palynomorphs are preferred. About ten sites and beds rich in organic matter gave sufficient material to support significant results. The main following interpretations may be drawn referring to five organic-rich intervals covering the entire region, in stratigraphic order, 1] a Louisiane type lanscape of uppermost Oligocene or lowermost Miocene age (Oret 606: Soyer, 1972; Russo Ermolli, 1991; Florennes CM zone I and Andenne Clay: Roche, 1998, unpublished), 2] more or less wet marshes with *Taxodiaceae* in the middle Miocene (Onhaye ECTP, Sosoye: Russo Ermolli, 1991 and Florennes CM zone II: Roche,1998, unpublished), 3] Maryland type marshes of (middle-) upper Miocene age (Bioul: Russo Ermolli, 1991; Fairon-Demaret,1994), 4] *Taxodiaceae* marshes disappear in the uppermost Miocene (Freyr: Soyer, 1972; Florennes CM zone III: Roche, 1998, unpublished), 5] *Sphagnum* grass land of early Pliocene (Florennes CB: Roche, 1998, unpubl.).These results indicate a long term karstic evolution during at least a part of the Miocene times.

Structure of the trapped sediments and karstic development

Calembert’s precise reports on the numerous clay mines acting in the Condroz during the first half of the 20th century provide unique geometrical data for understanding the structure of the cryptokarsts. No similar reports are available for the ESEM in spite of the evidence of some mining works (personal field observations). Nevertheless, some large quarries remained open until the eighties allowing local structural studies (Ertus, 1990; Lemy, 1996). Both evidence the same types of subsiding-related structures ranging between shallow entonnoirs with gently centripetal dipping and glove finger-like complex structures with vertical and overturned beds. The magnitude of the karstification rate is assumed to partly determine these structures. A moderate one resulted in a regular decrease of the dipping (80-90° to 20-10°) from the wall towards the centre as for the Champseau (Condroz) and Onhaye (ESEM) sites (Calembert, 1945; Perruchot *et al*., 1997). On the contrary, a high rate resulted in vertical and overturned bedding as shown in the Try-Dô-Baur, Vaudaigne or Manoux sites in Condroz (Calembert, 1945) or in the Bioul one in the ESEM (Ertus, 1990). Overturned structures may imply a certain imbalance between the subsiding-dissolution rate and the sedimentary supply.

As an example, the Weillen site, gives pertinent details of the development of such structures and on the behaviour of the sediments in the space freed by the dissolution (Fig.1). In this case, the dissolution could be related to a faulted zone in the limestone and the karst was forced to develop into a narrow elongated pocket (70-100m x 320m min.; Lemy,1996; Nicaise, 1998).The deformation of the infilling which is more often of “ductile” style, was brittle in this case. A sequence of faults developed offering to describe the relationships between the dissolution and the infilling of the pocket and allows to distinguish two main steps as reported in the figure 1.

In most of the karsts, the bedding thickness variations record an identical tendency in two main steps to which a last one can be added, *i.e.* in stratigraphic order and schematically: 1) ~10m of thin beds, more or less regular in thickness resting on the marine sands; 2) a few very thick beds, often organic-rich (Machuria) with large lateral thickness changes (~40-50 m to 0 m); 3) sometimes about 10 m of some regular thin beds (Not preserved in the quarries). We interpret the huge thickening of the second step as a result of an important increase of the accumulation/dissolution rate. The thickest beds are attributed to the Middle/Upper Miocene, so we can deduce that the karstic dissolution increases drastically during this times (~x5).

Weathering of the infilling and surrounding wall rock

The sediments accumulated in the cryptokarts exhibit different alteration degree. Sometimes the sediments appear very fresh as shown by the exceptional preservation of cellulose and hemicellulose in the wood pieces (De Putter *et al*.,1996). At the opposite, the carbonate wall rock is often silicified and halloysitised into several meters around the karsts (Ertus, 1990; Nicaise, 1998). The permeable sediments are regularly bleached and kaolinitised. Some minerals tend to disappear as felspars, certain clay and heavy minerals; the quartz grains are often corroded. Some others appear, quartz, halloysite (-kaolinite), iron and Mn oxides, and in few quantities, Al sulphates, gibbsite, monazite (Nicaise *et al*., 1996, unpublished data). The halloysite (-kaolinite) appears in large quantities (H1) in the early step of the evolution of the karst during the Lower Miocene in association with the quartz as *in situ* deposits replacing the limestone (Fig. 2). This early neogenesis of the halloysite explains why this deposit is often fractured and incorporated in a breccia association with silicified fragments. This mechanical reworking is induced by the dissolution-subsidence crisis above described (Fig. 2). Very small quantities of halloysite still formed at this time (H2).

We may assume that this major change in weathering processes was internally driven by a total exhaustion of mobile cations, Al, Si, K, Ca… from the sediments enhancing the aggressiveness of the percolating solutions. Another explanation could be proposed in relation with the history the nearby Rhine graben (Gliese and Hager, 1978), with which interesting correlations may be found with the fault activity and the shore shifts. The fault activity increases during the Upper Oligocene and the Lower Miocene. It then stabilizes until the Upper Miocene. So the development of the giant halloysitic cryptokarsts appears in very close coincidence with this phase of tectonic quiescence. The Upper Miocene also records a major regression which could be related with the end of the karstification.

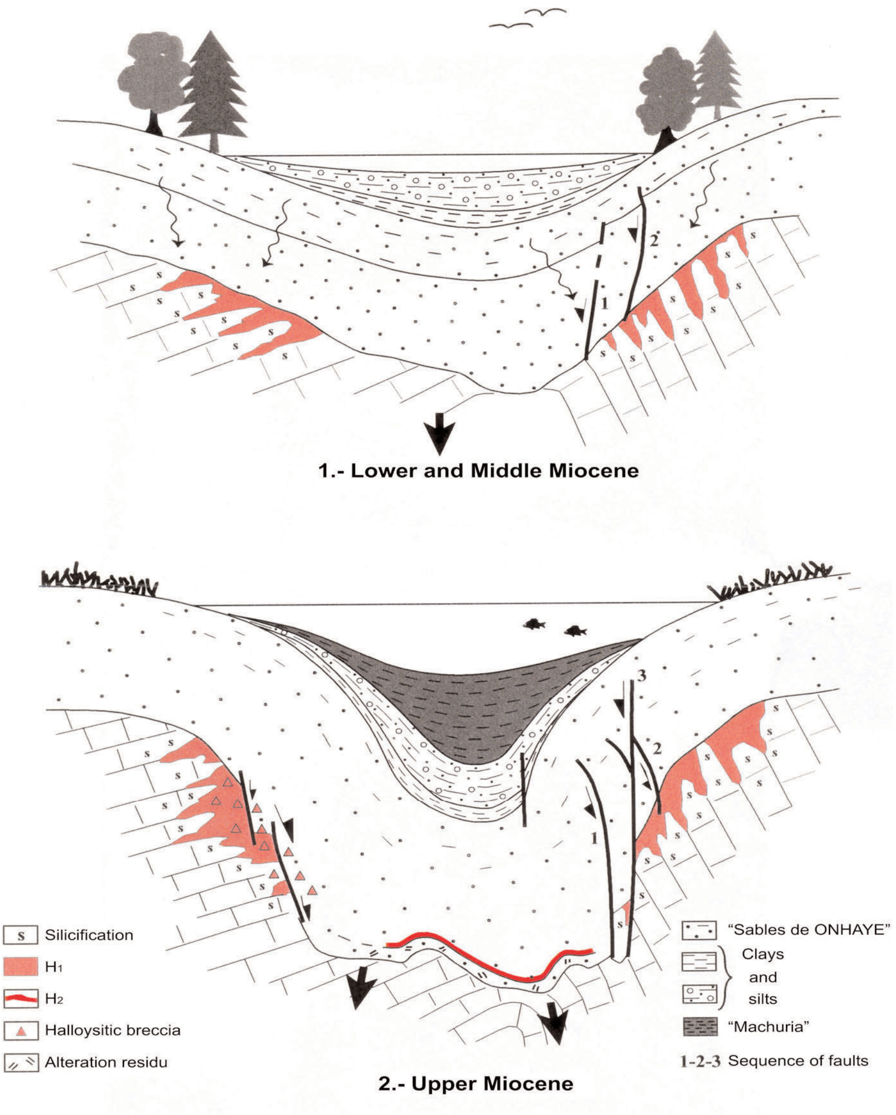

Fig. 1.- Main steps of the development of the karstic pockets of the ESEM-Condroz area. Schematic relationships of the weathering, evidenced by the halloysite occurrences, and the steps 1 and 2 of the karstic processes. Not to scale.

Model of paleoweathering processes and geochemistry

Chemical modeling of the Weillen karstic system was performed using the computer code Geochemist's Workbench (GWB) 3.2 (Bethke, no date). Such a modeling is *a priori* complicated by the relative monotony of the observed paragenesis, composed solely of silicates (kaolinite/halloysite, quartz), and of iron oxides (rare gibbsite). It is important to note that sulfates (and especially those sulfates allowing to constrain the pH range, as jarosite; see for instance De Putter *et al.*, 2000) were never found in significant amounts, in the karst.

The absence of gypsum (and of gypsum pseudomorphs) in the studied material is an important argument allowing to rule out any possibility that the karst deepening was the result of a downward percolation of highly acid (sulfuric) fluids produced through pyrite oxidation in the near-surface. Sulfuric fluids in carbonated environments should inevitably form gypsum, in much larger quantities (*i.e.* a factor $\sim 10^3$) than kaolinite.

The scenario that accounts for the larger quantity of observed data is the following: 1 liter of a basic meteoric fluid (see Stumm and Morgan, 1996) is allowed to react with 10 mg of fresh Grimmertingen Sands containing ~3% of pyrite, in the presence of atmospheric oxygen. The output of this model is shown in figure 2. All the minerals listed in figure 2 are present in the field. However, it is likely that this mineral indeed formed in the early phase of mineralization, when water activity was high enough (and silica activity low enough) to prevent kaolinite to form. Because thermodynamic parameters of halloysite are not included in the database, this mineral is not shown in figure 2. However, taking this mineral into consideration, the proposed scenario is still improved: gibbsite forms first, in high water activity media. It then converts to halloysite, with increasing silica activity (but still at constant water activity). And finally, halloysite is transformed into kaolinite when water activity decreases, for instance when the environment is better drained (Trolard *et al.*, 1990). The neoformation of quartz, at the end of the modeled reaction, appears to be in good accordance with the general occurrence of silicified limestone in the rim of the karst (Ertus, 1990; Nicaise, 1998).

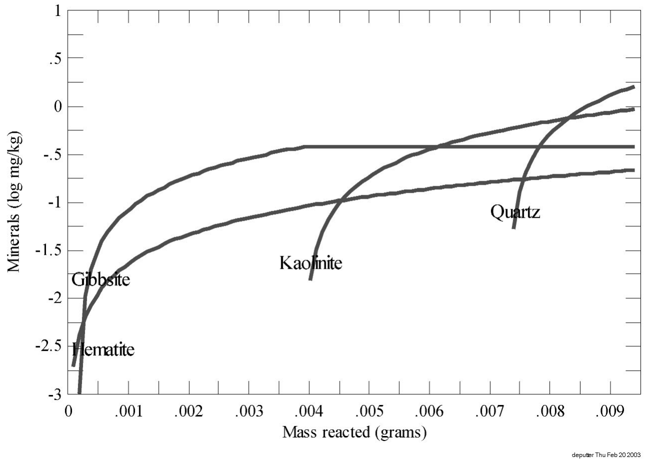

Fig. 2.- Chemical model output of the weathering paragenesis (Fe and Al oxides, halloysite-kaolinite, quartz), observed in the Weillen karst, using GWB 3.2.

Interestingly, in the above model, the limestone wall does not play an "active role" (chemically speaking) in the weathering and mineralization scenario. Rather, in our opinion, it acts as a mere mechanical support for the mineralization. This statement has potentially important consequences for the chemical "budget" of some major cations, within the system. For instance, Ca^{2+} is mobilized when limestone dissolves during the karst deepening, but it does not appear to be trapped within any neoformed mineral. This means that the proposed karstification scenario should allow for a major Ca^{2+}-loss.

The figure 2 output shows that halloysite/kaolinite formed at a rate of ~1mg/liter acidic fluids, *i.e.* with a fluid/rock ratio of ~10^6. As previously stated, this suggests that ~5.10^{10} l of meteoric fluid were necessary to form the ~5.10^4 tons of clays in the Weillen karst (De Putter *et al.*, 2002). Such a fluid quantity appears to be a rather low figure, if the mineralization process was actually efficient during most of the Middle/Upper Miocene periods, *i.e.* ~ 10 Ma (Nicaise, 1998). Further work is needed to decide whether the weathering process was of low intensity and long duration, or rather of high intensity (which is not suggested by the above modeling) and short duration, though most available data would support the first hypothesis.

Concluding remarks

Neogene cryptokarsts of Ardenne exhibit an unusual large development that can be explained through chronological and geochemical arguments. Both plaid for a long evolution encompassing at least a part of the Miocene. Tectonic control of triggering, evolution and stopping of the karstification seems to be a powerful way to explain the main specificities of these karsts but it needs further investigations. In the near future, a special attention will be devoted to the European structural context during the Miocene.

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