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Diagenetic and palaeogeographic significance of clay, carbonate and other sedimentary components in the middle Devonian limestones of western Ardenne, France

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Abstract

Givetian deposits accumulated in a more than 300-m-thick succession on a carbonate platform developed on the passive margin of the so-called Rheic ocean. These rocks crop out in the Ardenne massif at Glageon in a structural context marked by small fault-controlled basins subjected to moderate overburden, tectonic and metamorphic constraints. Data on the clay mineral distribution, associated with detailed information on limestone facies/microfacies, as well as data on the depositional profile and sequence stratigraphic evolution of these rocks indicate strong carbonate diagenesis and moderate clay diagenesis, the latter being favoured by the early occurrence of the former, which prevented further fluid–rock interaction. The clay assemblage distribution, which roughly parallels the palaeomorphologic and palaeobathymetric shape of the Givetian continental margin, is organized in five successive zones indicating large-scale sequence stratigraphic evolution and the control of regional sea-level fluctuations. The clay and other sedimentary components provide additional information on the warm and variably humid climate, northern, relative to southern, terrigenous sources, the open sea relative to restricted depositional environments, the temporary tectonic rejuvenation of the continental hinterland and the chemical conditions allowing early diagenetic modifications.
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1. Introduction

1.1. Purpose

A more than 300-m-thick succession of Givetian (Middle Devonian) platform limestones crops out at Glageon in the western part of the Ardenne massif close to the France/Belgium border. As this outcrop is located in a moderately tectonized and unmetamorphized area and is well-known from both lithological and tectonic points of view (e.g. Boulvain et al., 1994), we have considered it as a potential case study for delivering new information on both paleoenvironmental and diagenetic successions

The aim of the present paper is to inter-relate data on carbonate lithofacies, microfacies, clay mineral distribution and sequence stratigraphy in order to better identify the respective paleoenviron-

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mental and diagenetic constraints on the formation of Givetian limestones and to further characterize the palaeogeography of western Europe in Middle Devonian times.

1.2. Geological and structural context

The Variscan margin in Belgium and northern France is represented by Brabant parautochthonous and Ardenne allochthonous massifs (Fig. 1). Both are constituted from early Paleozoic (Cambrian, Silurian) incompetent basement, rich in siliciclastic formations which have been subjected to Caledonian–Acadian deformation (Michot, 1976; Meilliez and Mansy, 1990; Meilliez et al., 1991; Mansy and Meilliez, 1993). The subsequent Paleozoic sedimentation on the passive margin of Ardenne developed during Devonian time in small fault-controlled basins which successively formed from south to north. The sediment accumulation in these basins was higher overall to the south, with strong variations close to some fault systems, suggesting tectonic control during most of the Devonian period. Extensional tectonics occurred close to the Rocroi massif, as indicated by the Middle–Late Devonian intrusion of granodiorite dykes which was followed by low grade metamorphism (Goffette et al., 1991).

At the end of the Early Devonian (Late Emsian) and during the Middle and Late Devonian,

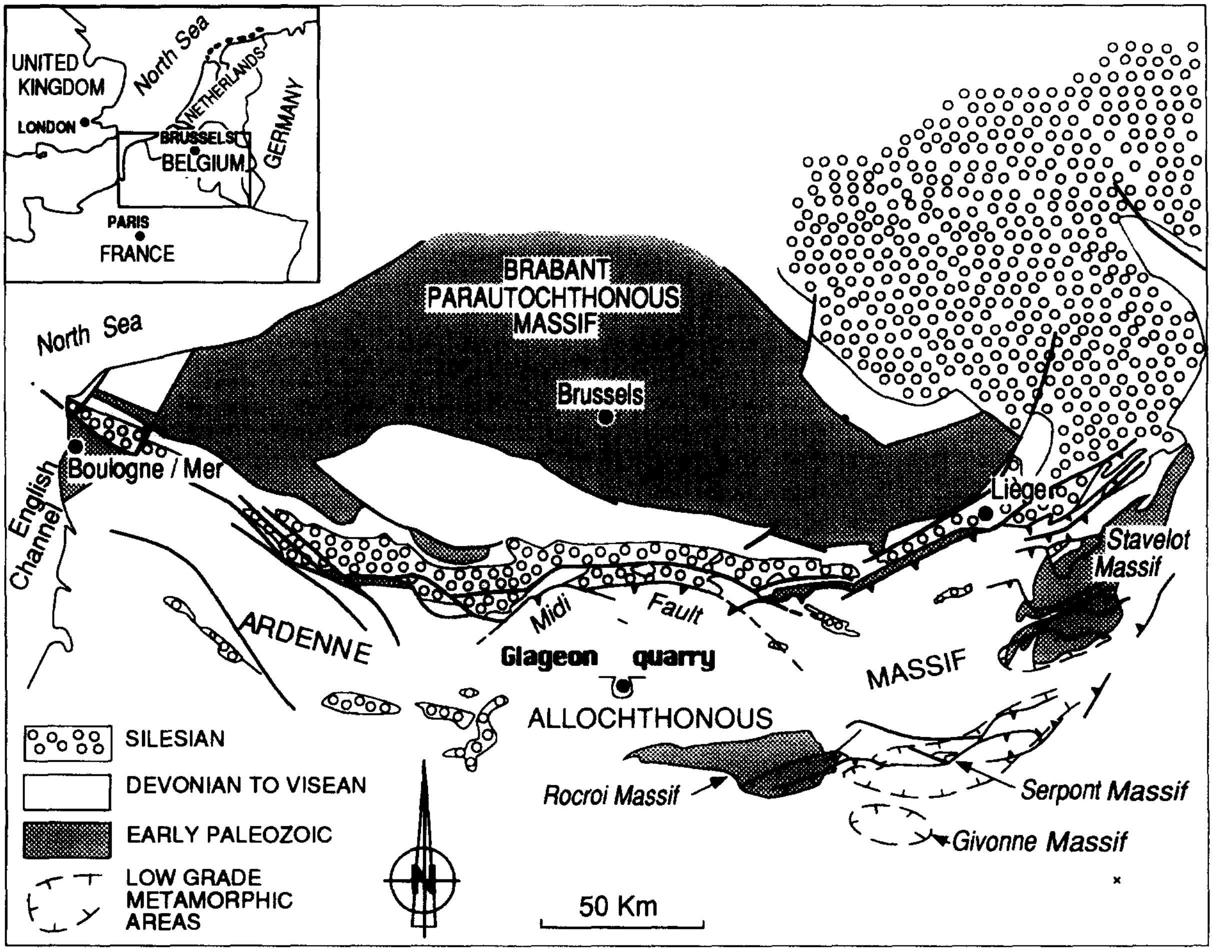


Fig. 1. Location of the Glageon quarry and geological-structural framework.

platform carbonates and associated evaporites were deposited along the northern margin of the Ardenne in shallow-water environments, whereas mud and some volcanics accumulated in deeper environments to the east and southeast. Carbonate deposits were enriched in clastic components and associated with reefs during the Frasnian (Late Devonian), relative to the Givetian (Middle Devonian).

1.3. The Givetian section at Glageon

The Givetian platform limestones, which constitute the upper part of the Middle Devonian formations, crop out widely along a 200-km-long belt in the Ardenne massif at the northern France–southern Belgium transition where the Givet type-area is located (Préat and Mamet, 1989). These formations belong to the Dinant synclinorium which results from a Carboniferous structural episode that includes northward to northwestward tectonic transport and progressive deformation, with successive Visean to Westphalian development of ramps, folds and faults. The overburden is estimated to have never exceeded 2 km (Mansy and Meilliez, 1993; Mansy et al., 1995).

The Glageon quarry is located in northern France within the Avesnois region, west of the well-known Givetian outcrops at the southern border of the Ardenne allochthonous massif (Fig. 1). It contains the five uppermost Eifelian–Givetian carbonate formations classically described in the Ardenne and termed Hanonet, Trois-Fontaines, Terres d'Haurs, Mont d'Haurs and Fromelennes (see Préat and Mamet, 1989). This quarry has been subjected to detailed investigations (Boulvain et al., 1994) because (1) the Givetian series is nicely exposed in a thickness exceeding 300 m, (2) the structural deformation is moderate and (3) there is an absence of noticeable sedimentary gaps or section duplication suggesting almost continuous deposition. The Glageon section is located on a wide staircase fold with a limb dipping at 50° N. Pressure solution cleavage is penetrative throughout the section and follows a previous phase of stylolithisation. Slickensides in the bedding plane are numerous and open fractures occur frequently, but only small faults can be identified.

1.4. Sampling, methods and objectives

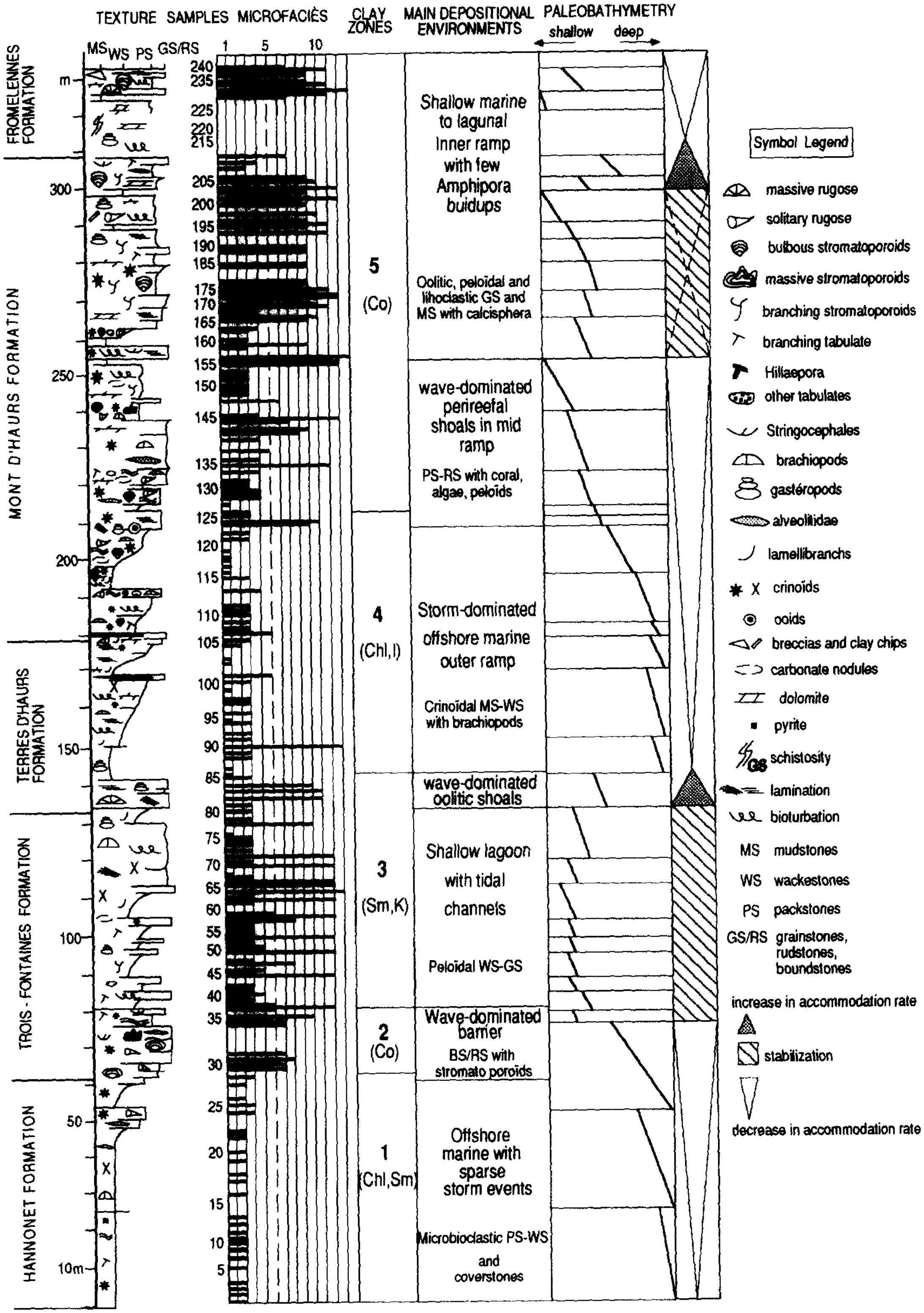
We have described and extensively sampled the five uppermost Eifelian to Givetian formations outcropping on a 330-m-thick succession in the Glageon quarry and have performed high-resolution investigations of carbonate lithofacies and microfacies for the whole section. A total of 240 petrographic thin sections were described. Semi-quantitative estimates of the abundance of more than 20 biologic and mineral constituents were used for the identification of carbonate microfacies (see also Boulvain et al., 1994).

The clay fraction of Glageon limestones constitutes usually less than 5% of the bulk rock. Seventy-five samples, regularly spaced along the 300 m of section, have been investigated by X-ray diffraction using oriented mounts of the less than 2 μm size fraction containing non-calcareous particles (see Holtzapffel, 1985). A few select samples, corresponding to the main clay mineral facies have been observed by transmission electron microscopy or subjected to inorganic geochemical analysis (atomic absorption spectrometry).

2. Facies description and interpretation

Thirteen main carbonate microfacies were identified along the Glageon section (see 3rd column in Fig. 2). According to their texture and biologic content (Wilson, 1975), they are successively described from open marine to restricted environments. Each facies description is followed by an interpretation of its depositional environnement. The interpretation is based on analogies with present-day environments and comparisons with ancient carbonate platform models (Wilson, 1975; Armstrong and Mamet, 1977; Préat and Boulvain, 1982; Préat and Mamet, 1989).

Facies 1. *Clay bearing mudstones and wackestones with brachiopods and crinoids.* This facies locally includes some pelecypods, trilobites and ostracods. Deformation due to bioturbation is often important and pyrite is frequent. The abun-


TEXTURE
SAMPLES
MICROFACIÈS
CLAY ZONES
MAIN DEPOSITIONAL ENVIRONMENTS
PALEOBATHYMETRY
shallow
deep
MS WS PS GS/RS
1 5 10
FROMELENNES FORMATION
MONT D'HAURS FORMATION
TERRES D'HAURS FORMATION
TROIS - FONTAINES FORMATION
HANNONET FORMATION
m
300
250
200
150
100
50
10m
240
235
225
220
215
205
200
195
190
185
175
170
165
160
155
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95
90
85
80
75
70
65
60
55
50
45
40
35
30
25
20
15
10
5
5
(Co)
4
(Chl,I)
3
(Sm,K)
2
(Co)
1
(Chl,Sm)
Shallow marine to lagunal Inner ramp with few Amphipora buidups
Oolitic, peloïdal and lihoclastic GS and MS with calcisphera
wave-dominated perireefal shoals in mid ramp
PS-RS with coral, algae, peloïds
Storm-dominated offshore marine outer ramp
Crinoïdal MS-WS with brachiopods
wave-dominated oolitic shoals
Shallow lagoon with tidal channels
Peloïdal WS-GS
Wave-dominated barrier
BS/RS with stromato poroïds
Offshore marine with sparse storm events
Microbioclastic PS-WS and coverstones
Symbol Legend
massive rugose
solitary rugose
bulbous stromatoporoids
massive stromatoporoids
branching stromatoporoids
branching tabulate
Hillaepora
other tabulates
Stringocephales
brachiopods
gastéropods
alveolitidae
lamellibranchs
X crinoïds
ooids
breccias and clay chips
carbonate nodules
dolomite
pyrite
GS schistosity
lamination
bioturbation
MS mudstones
WS wackestones
PS packstones
GS/RS grainstones, rudstones, boundstones
increase in accommodation rate
stabilization
decrease in accommodation rate

dance and good preservation of open-sea organisms, the massive sediment texture and the fairly high amount of clayey material, as well as the scarcity of algae, indicate an open marine environment probably located beneath the base of wave action.

Facies 2. *Microbioclastic packstones and coverstones* (Tsien, 1981) *with lamellar stromatoporoids and corals (alveolitids, coenitids)*. The microbioclasts are generally oriented and a rough lamination occurs locally. Other bioclastic components are recognized, such as crinoids, ostracods, trilobites, issinellids, proninellids, kamaenids, brachiopods, tabulate and solitary rugose corals, pelecypods and some bryozoan (fenestellids). Bioclasts are often affected by bioerosion and deformation due to bioturbation is intense. The sediment is bound by lamellar frame builders. This facies is typical of pre-reefal or peri-reefal environments, moderately agitated by waves, where lamellar organisms play a pioneer role in the establishment of coelenterate-dominated bioconstructions.

Facies 3. *Wackestones and packstones with peloids, crinoids, algae (paleosiphonocladales) and gastropods*. Some brachiopods, bryozoans, ostracods and trilobites occur locally. Issinellids often form some bioclastic lenses with rough lamination. Grains are usually micritized and bio-eroded. Asymmetrical coatings occur frequently. These sediments accumulated close to issinellid-forming bioconstructions in an open marine, temporarily agitated environment located close to permanent wave base.

Facies 4. *Rudstones with corals (solitary rugose, branching and nodular tabulate), crinoids and brachiopods*. The matrix is locally argillaceous and includes some pelecypod fragments, paleosiphonocladales and bryozoans. This faunal assemblage is attributed to peri-reefal environments dominated by clastic contributions issuing from adjacent bioconstructions.

Facies 5. *Peloidal and crinoidal grainstones*. Relatively well sorted, the facies includes some brachiopods, gastropods and micritized grains. It characterizes a high-energy, shallow marine environment located in an open marine setting (Wilson, 1975)

Facies 6. *Bindstones and rudstones with irregular massive stromatoporoids, corals (tabulate, rugose) and crinoids*. The spaces between the sometimes meter-scale stromatoporoids are filled with crinoids, pseudo-algae (issinellids, proninellids, kamaenids) and peloids. An encrusting by frame-builder organisms (e.g., alveolitids–stromatoporoids) is frequent, but algal-bacterial coatings occur occasionally (*Girvanella, Bevocastria, Sphaerocodium*). Because the frame-builders constitute large biostromal accumulations stabilized by encrusting algae, the depositional environment is considered turbulent, shallow and located in the permanent wave zone. The numerous coatings indicate a relatively high biogenic competitiveness typical of reef environments (Fagerström, 1987).

Facies 7. *Floatstones with corals, algae (paleosiphonocladales) and peloids*. Clasts of frame-builders are essentially associated with issinellids and peloids. Micritisation of grains, as well as symmetrical algal coatings, occur frequently. These sediments probably formed in a shallow marine, back-reef environment with constant and moderate agitation. A relatively low rate of sedimentation allowed the development of micritic and algal coatings.

Facies 8. *Branching stromatoporoids rudstones*. The branching stromatoporoids are mainly represented by Amphipora and accompanied by a few peloids, pelecypod and algae (*Girvanella*, Codiaceae) fragments. These accumulations result from the reworking of oligospecific bio-constructions in a rather restricted marine environment that was very shallow and only temporarily submitted to wave agitation.

Facies 9. *Packstones and grainstones with algae (paleosiphonocladales, Codiacea), Calcisphera and peloids*. Few gastropods and pelecypods are iden-

Fig. 2. Formations, textures and main components, microfacies designation, clay-zone distribution, broad classes of depositional environment, paleobathymetric variations and sequence stratigraphic interpretation in the uppermost Eifelian–Givetian limestones of Glageon quarry. Microfacies numbers refer to the facies described in the text. Clay zones: *Co* = corrensite; *Chl* = chlorite; *I* = illite; *Sm* = smectite; *K* = kaolinite.

tified. Both Codiaceae and *Girvanella* form symmetrical coatings. These sediments result from the reworking of intra-lagoonal bio-constructions with issinellids and paleoberesellids, and correspond to an only slightly agitated environment.

Facies 10. *Oolitic grainstones*. Proto-oolitic peloids are locally abundant. Some paleosiphonocladales, gastropods, micritized grains, crinoids and coral fragments occur locally. The grain-size is very uniform. The cement consists of drusy sparite. These grainstones are typical of constantly agitated, but restricted environments which could be referred to as intra-lagoonal bars, possibly in connection with tidal channels.

Facies 11. *Grainstones with peloids, proto-ooids and lithoclasts*. The millimeter- to multimillimeter-sized lithoclasts exhibit an irregular shape. They are often associated with gastropods, oolites, sparse algae (*Girvanella*, Codiaceae, paleosiphonocladales, Udoteaceae), crinoids, brachiopods and coral fragments. The sediment is generally well-sorted and sometimes exhibits planar and cross-laminations. The lithoclasts result from the reworking of intra-lagoonal and/or temporarily-exposed intertidal deposits. This facies is attribued to channel and shallow pool environments (Wilson, 1975).

Facies 12. *Calcisphere-bearing mudstones*. These mudstones are associated with some *Bisphaera* as well as with infrequent pseudo-algae (proninellids, kamaenids) and gastropods. They suggest a deposition in a lagoonal, strongly restricted environment, characterized by a very low biological diversity.

Facies 13. *Laminar mudstones*. Mudstones locally enriched in microbioclasts. Dolomitisation is frequent and pyrite is locally abundant. The laminations could have been caused by tide or storm action in sediment deposited close to exposure depths under sub-evaporitic conditions (Boulvain and Préat, 1986).

The diagenetic history of middle Devonian carbonates of the Glageon region has been investigated by petrographic and geochemical studies (Boulvain et al., 1992). Cathodoluminescence analyses of carbonate cements from grainstones and fenestrae reveal that the bulk material is composed of xenomorphic, granular, dull orange, luminescent calcite. This cement developed in the cavities and between the grains through a fracture network contemporaneous with pressure-solution phenomena. The calcite is very homogeneous and rarely zoned. Its very low $\delta^{18}O$ values together with its petrographic characteristics and iron content point to a late diagenetic origin.

3. Paleobathymetric trends, depositional profiles and sequence stratigraphy analysis

Some of the carbonate microfacies identified are present throughout the Glageon section, whereas others are restricted to either the Hanonet and Trois-Fontaines Formations, or the Terres d'Haurs, Mont d'Haurs and Fromelennes Formations. For example microfacies (2) and (6) occur only in the lower part of the section, and microfacies (4), (8) and (10) only in the upper part. Due to this occurrence of mutually exclusive microfacies, two distinct depositional profiles are proposed to explain the distribution and the evolution of the paleoenvironments (Fig. 3).

The lower part of the section, which includes the Hanonet and the Trois-Fontaines Formations, exhibits from base to top the following succession (Figs. 2 and 3A): (a) storm-influenced, offshore marine, microbioclastic packstones–wackestones and then reef sole coverstones at the toe of a prograding barrier front (facies 2); (b) Stromatoporoid-rich, bindstone–rudstone of a wave-dominated reefal barrier (facies 6); and (c) relatively restricted marine, peloidal packstones–grainstones characterizing a back-barrier depositional environment (facies 7, 11). This microfacies succession closely resembles the paleoenvironmental distribution observed along asymmetrical profiles of carbonate platforms marked by a barrier at the shelf-slope break (Wilson, 1975; Read, 1985) (Fig. 3B).

In contrast, the upper part of the section including the Terres d'Haurs, Monts d'Haurs and Fromelennes Formations, exhibits from base to top (Figs. 2 and 3B): (a) crinoidal mudstones–wackestones with brachiopods in a deep marine, storm-dominated environment (interbedding of facies 1 and 3); (b) cross-bedded packstones–

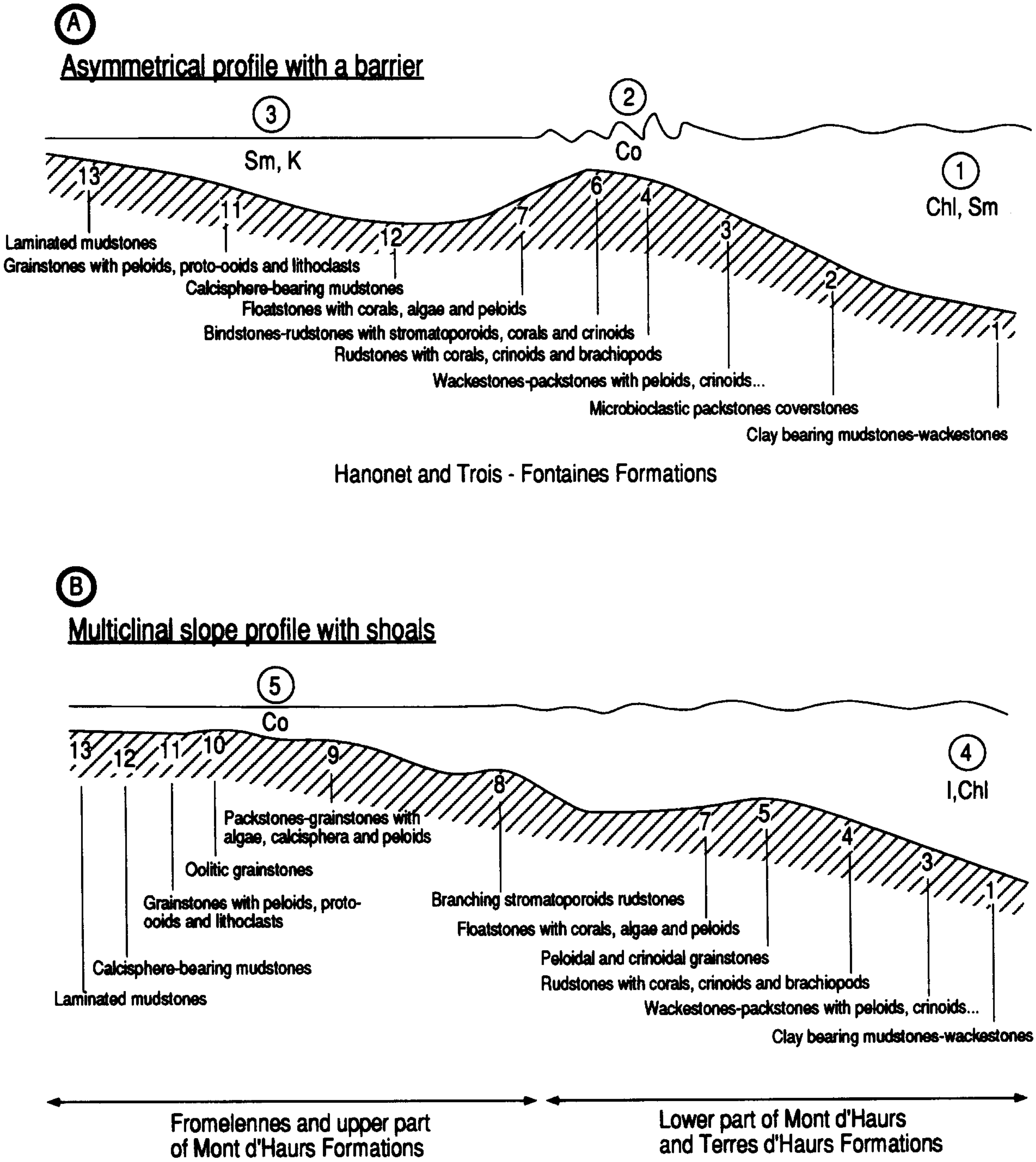

Fig. 3. Depositional models identified in the Glageon section and major trends of clay mineral variations. The arabic numbers refer to the carbonate facies described in the text. (A) Lower part of the section: paleoenvironnemental distribution along an asymmetric profile with a barrier (facies 6) and a barrier front (facies 2). (B) Upper part of the section: repeated transition from open marine to restricted environments. Clay minerals: same symbols as in Fig. 2. The circled numbers correspond to clay mineral zones (see text).

rudstones with highly diversified fauna (corals, algae) and peloids interpreted as corresponding to wave-dominated shoals (facies 4, 5, 7 and 8); and (c) calcisphere-bearing mudstones and oolitic, peloidal and lithoclastic grainstones with sparse occurrences of branching stromatoporoid buildups attributed to shallow marine, restricted lagoonal deposits (facies 9, 10, 11, 12, 13). This succession does not exhibit any physiographic break similar to the barrier observed in the Trois-Fontaines Formation. The Terres d'Haurs, Monts d'Haurs et Fromelennes Formations appear to have been organized along a multiclinal slope profile (Fig. 3B) which experienced a progressive landward restriction controlled by a ramp shelf (Ahr, 1973; Burchette and Wright, 1992).

These two facies successions correspond to broad shallowing upward trends from deep, open marine to shallow, restricted marine depositional environments bounded by thin deepening upward trends, as is the wave-dominated oolitic shoal belt located at 130 m in the cross-section (Fig. 2). They could be considered as Maximum Flooding Surface (MFS)-bounded depositional sequences (Galloway, 1989). On a smaller scale, the two depositional sequences are made up of 5–10-m-thick sediment packages (Fig. 2) that exhibit a rough shallowing upward trend and appear to fit the parasequence definition (Van Wagoner et al., 1988). Into each depositional sequence, the vertically-stacked parasequences build up progradational, agradational and retrogradational parasequence sets. The progradational–agradational stacking patterns exhibit a progressive facies drift from deep to shallow marine environments, with a poorly defined thickening-upward tendancy and an upsection increase in the number of erosion and exposure surfaces along with the shallowing-up character that typifies a decreasing rate of accommodation. The retrograde parasequence sets exhibit a decrease in the number of exposure surfaces progressively replaced by flooding surfaces that typify an increasing rate of accommodation. In the second sequence (Fig. 3b), the agradational parasequence sets are slightly retrogradational and then progradational. This may characterize a subdued sequence in a lowstand position, and needs to be checked by correlations with other sections on a regional scale. At the scale of the whole section at Glageon, the two depositional sequences identified in Fig. 3 characterize a shallowing-upward trend expressed by increasing-upward restriction in lagoonal environments and intense dolomitisation as observed in the upper Mont d'Haurs and lower Fromelennes Formations.

4. Clay mineral distribution and diagenesis

4.1. Results

In spite of its small abundance (from about 1–5% of the bulk rock), the clay fraction in Glageon Givetian limestones is characterized by an unexpected diversity of minerals and strong depth variations in their abundance (Fig. 4). The clay minerals include some common species such as chlorite (traces to 35% of the clay fraction), mica-illite (30–90%), fairly crystalline smectite (0–45%), kaolinite (0–10%), random mixed-layers comprising illite-, chlorite-, smectite- and/or vermiculite-layer types (0–5%), and also less usual minerals such as subregular chlorite–smectite mixed-layers [i.e., corrensite sensu lato (s.l.); abundance varying from 0 to 65% of the total clay minerals]. Associated non-clay minerals comprise the following species: ubiquitous, rare to common quartz; frequent, rare to common feldspars; occasional, rare to abundant fluorite and very rare and occasional goethite or opal. Notice that a similar diversity of clay assemblages has been recently identified in Eifelian and Givetian limestones of Remonchamps section, located in the northeastern part of the Dinant basin in Belgium (Yans, 1995).

The vertical distribution of the mineral species in the clay fraction of Glageon limestones is not random, allowing the identification of five successive clay mineral zones from bottom to top of the section (Figs. 4 and 5):

– Zone 1 (0–62.6 m) is characterized by fairly abundant chlorite, the percentage of which tends to decrease upsection from underlying Eifelian limestones to early Givetian deposits (Hanonet Formation). Chlorite is associated with common to abundant feldspars, and little but constantly

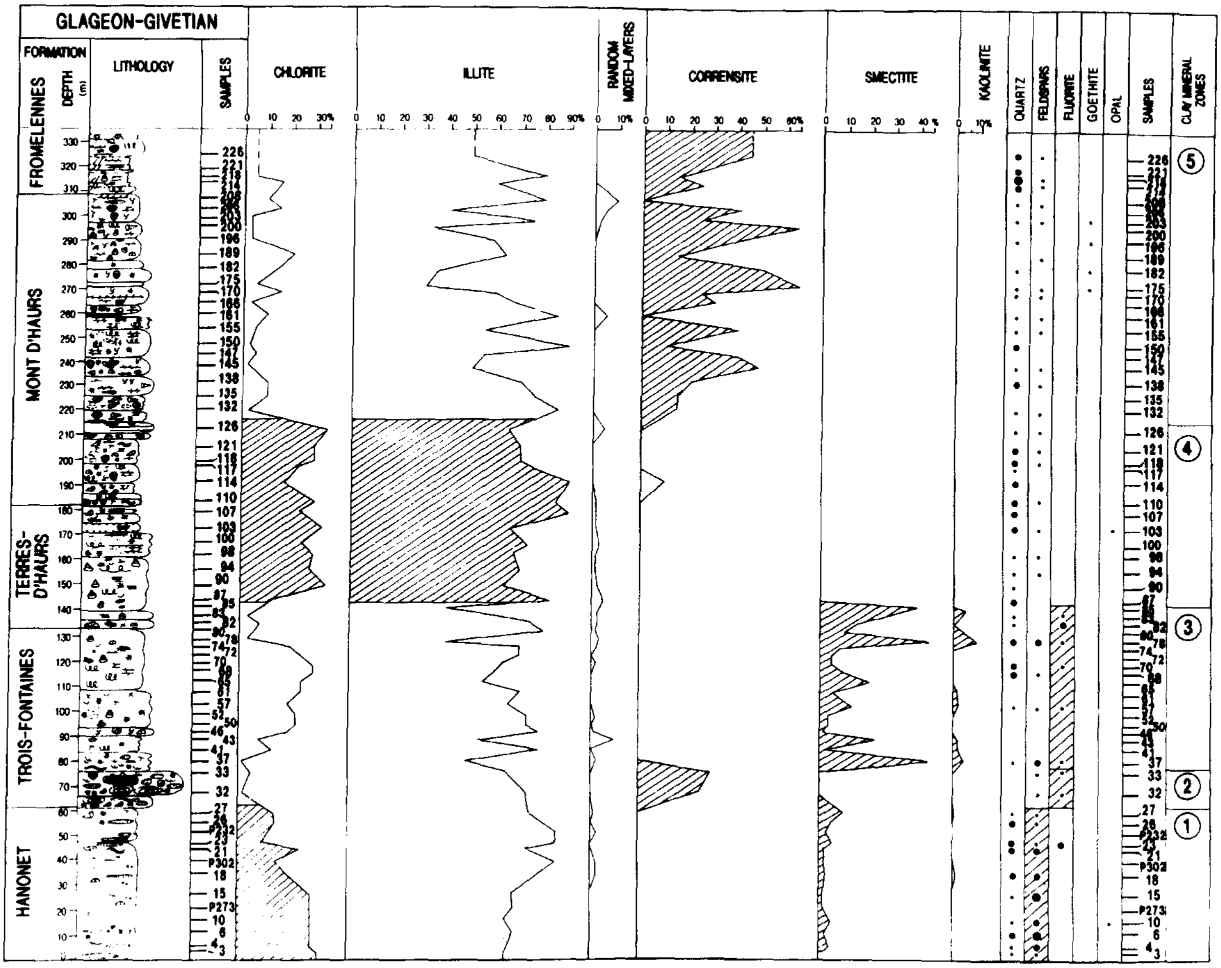

Fig. 4. Mineralogy of the clay fraction. Facies legend: see Fig. 2. The shaded zones correspond to significant clay mineral characteristics and associated minerals. Non-clay minerals: common = big dot; little abundant = medium-sized dot; rare = small dot.

present smectite. Illite is abundant as in most other parts of the section.

- Zone 2 (62.6–80 m) correlates with the biostrome (facies 6) marking the lower part of Trois-Fontaines Formation. Of moderate thickness, it contains fairly abundant corrensite (25–30% of the clay fraction) associated with fluorite.

- Zone 3 (80–142 m) has a large depth range, it covers most of Trois-Fontaines Formation and the base of Terre d'Haurs Formation. Zone 3 is characterized by variable, sometimes large amounts of smectite (5–45%), as well as the presence of significant amounts of kaolinite (up to 10%) and fluorite. Chlorite and especially illite are usually less abundant than in other clay mineral zones (15 and 55% against 10–25 and 58–75%, respectively; see Fig. 5).

- Zone 4 (142–215 m) occupies the largest part of Terre d'Haurs Formation and the lowest third of Mont d'Haurs Formation. The rather stable clay association strongly differs from both underlying and overlying clay zones. It is marked by abundant, well crystalline chlorite (10–35%) and illite (65–90%). Other minerals are absent or very rare, except quartz (rare to common) and feldspars (rare).

- Zone 5 (215–330 m) has a large depth range,

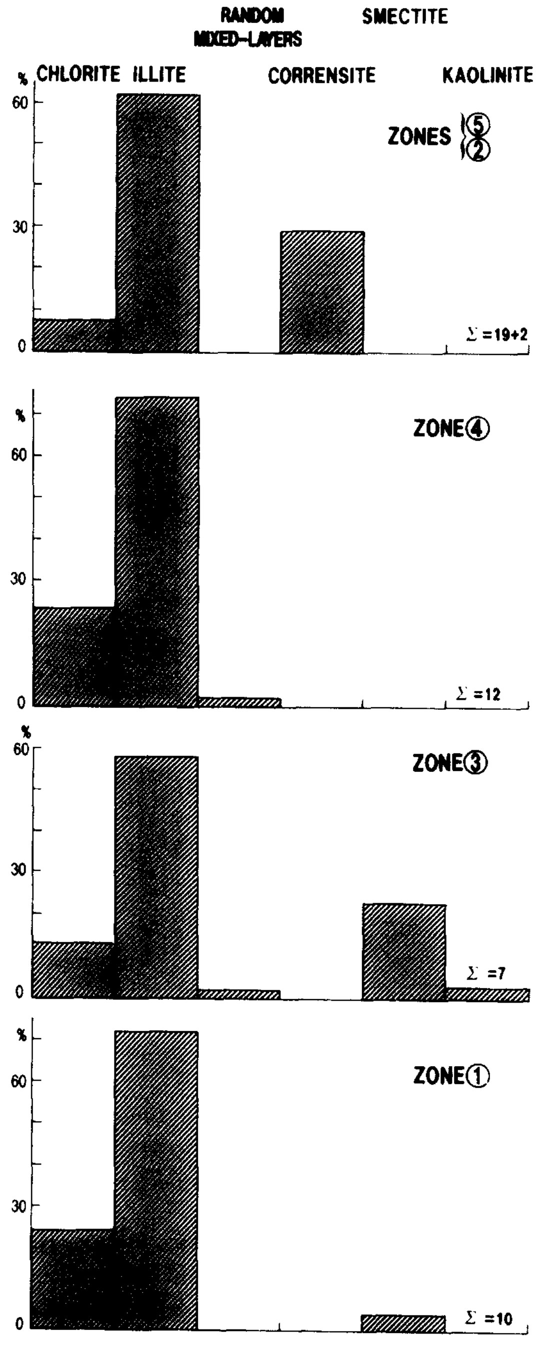

Fig. 5. Average clay mineral abundances in clay mineral zones in the Glageon section. $\sum$ = number of samples.

it covers most of Mont d'Haurs formation and the outcropping part of the Fromelennes Formation. The clay fraction displays an uncommon mineral assemblage characterized by corrensite (10–65%), which is not accompanied by smectite or fluorite. Chlorite and illite have here the lowest percentages in the whole section, except at levels where corrensite is less abundant.

4.2. *Clay diagenesis*

Control by overburden and tectonics. Smectite, kaolinite and random mixed-layers constitute clay minerals which commonly form at surface conditions during weathering and soil-forming processes (Millot, 1970; Chamley, 1989; Weaver, 1989). Their persistence, especially of smectite, in the detrital clay mineral assemblage in Glageon limestones suggests that the clay diagenesis, driven by burial effects, was probably of moderate, or even low intensity since the end of Givetian about 380 m.y. ago. The lack of any continuous change in clay mineral composition from the top to the bottom of the 330-m-thick section at Glageon supports such an interpretation (e.g., Kisch, 1983). Smectite reaches values as high as 45% in the clay zone 3 located just beneath the richest chlorite- and illite-bearing levels (zone 4). It should be noted that smectite is also present in the 300-m-thick Eifelian limestones cropping out at Glageon below Givetian formations (pers. unpubl. analyses). Most litho- and micro-facies at Glageon correspond to massive fossil-bearing limestones which are very little permeable to fluid migrations. This probably prevented the modifications of detrital smectite, as already pointed out in a few other diagenetic environments (e.g., Wilson et al., 1968; Bouquillon et al., 1985). It is significant that at Glageon, most of the clay mineral changes identified occur independently of the formations succeeding one another from Trois-Fontaines to Fromelennes during the Givetian stage (Fig. 4). The presence of subregular chlorite–smectite mixed-layers (corrensite) in the upper part of the section (zone 5) and their decrease in the underlying zone 4 at the benefit of chlorite could express an effect of the burial depth, but such a possibility is refuted by the appearance of smectite and of sub-regular mixed-layers in underlying clay mineral zones 3, 2 and 1. The illite crystallinity (width of the 10 Å XRD peak at half-height) usually shows low values (3–4.5 mm) which are a priori compatible with anchizone burial effects (Kisch, 1983); but the absence of a continuous vertical trend and the coexistence of illite with surface-derived minerals (smectite, kaolinite,) suggest that the former species is mainly detrital and was actively eroded from mica-rich, relatively unweathered substrates (see Chamley, 1989).

These results for both the diversity of the clay minerals identified and the non-gradational vertical distribution of their assemblages suggest that burial diagenesis and metamorphic effects linked to the tectonic effects have been of minor intensity in the Middle Devonian series investigated at Glageon in the southern part of the western Ardenne massif (Fig. 1). Such a deduction is a priori unexpected for sedimentary series as old as 375 m.y., but they have been only locally subjected to moderate lithostatic overburden (about 2 km) and tectonic constraints linked to the Hercynian orogenesis (Meilliez et al., 1991).

Several other data indicate that the Glageon area has been protected from important clay mineral diagenetic evolution. The sedimentary overburden seems to have been thinner than in other Ardenne regions, as suggested by the colour alteration index (CAI) of conodonts, the values of which increase when the geothermal gradient increases (Epstein and Harris, 1977). Helsen (1992) has measured remarkably low CAI values close to 3.0–3.5 (i.e. about 100–150°C), on the northern flank of Rocroi massif where Glageon area is located, whereas the other areas display values reaching 4.5–5.0 (250–300°C) (Fig. 6) and even 5.5 (more than 300°C) to the south of Rocroi massif. The low values registered close to Glageon indicate that the sedimentary overburden probably never exceeded 2000 m, a thickness resulting in rather short heating events and insufficient to allow the burial diagenesis to cause important clay mineral changes (Kisch, 1983; Chamley, 1989). In addition the time during which the lithostatic load in the Glageon area attained values of about 2 km is estimated at only 30 m.y. (Helsen, 1995), which prevented long-lasting diagenetic conditions. This is due to the fact that the western Ardenne region experienced tectonic uplift as early as the lower Carboniferous, which precluded subsequent thick and deep-water marine sedimentation.

The post-Givetian metamorphic events started at about 372 m.y. in Brabant parautochthonous massif (André et al., 1981) which is situated to the north of the study area (Fig. 1). South of the study area and in the Rocroi massif, the metamorphism started later and showed different phases culminating in greenschist, partially retrogradate conditions, from 450 to 350–300°C, with an estimated pressure of 3–2 Kb (Potdevin and Goffette, 1991). In contrast, to the north of the Rocroi massif towards the southern side of the major syncline constituting the Ardenne allochthonous massif, the metamorphic activity decreased, toward the Glageon area resulting in an especially low grade metamorphic zone. As a result the clay mineral assemblages in this area probably experienced only moderate diagenetic effects, which were unable to achieve anchizonal characteristics.

The Givetian limestones, the carbonate fraction of which has suffered strong diagenetic recrystallization (Boulvain et al., 1994), nevertheless have oxygen isotope characteristics indicating exposure to only mild thermal conditions (i.e. less than 120°C acting during less than 30 m.y.) in several quarries located on the southern flank of the Dinant synclinorium (Weiss and Préat, 1994; Helsen, 1995), including the pre-Givetian (Eifelian) limestones at Glageon (A. Préat, pers. commun.). This suggests that the carbonate diagenesis was developed in the early stages of burial and deformation, inducing early carbonate crystallization and reduction of the limestone porosity, therefore preventing subsequent fluid migration and water–rock interaction. This early reduction of porosity preserved initial characteristics of the clay minerals scattered in the limestones, which could explain the presence at Glageon of smectite and random mixed-layers. As a consequence, smectite and associated clay minerals at Glageon are considered to represent for the most part the remains of the detrital minerals deposited on the carbonate platform developed on the Ardenne continental margin during Givetian time. The clay minerals may therefore help to reconstruct the paleoenvironmental conditions existing in Givetian time, as is the case for smectites in other low

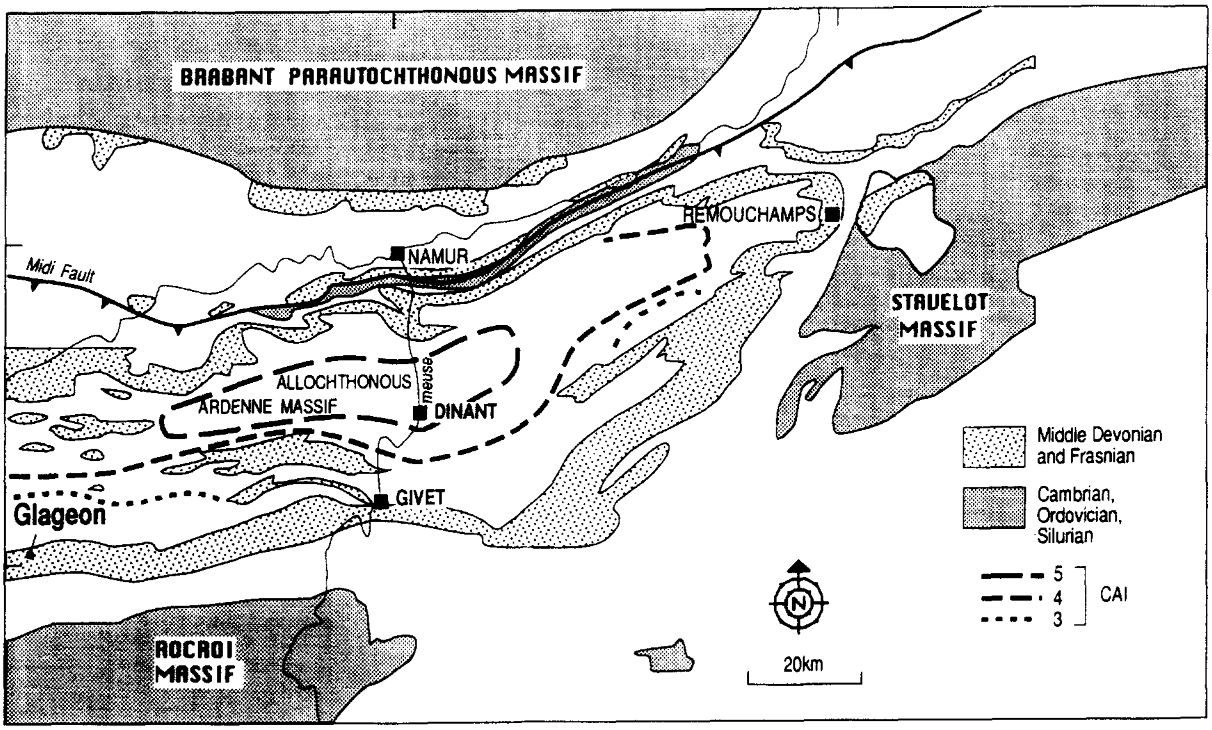

Fig. 6. Close-up of the geological map of the western Ardennes and colour alteration values of Conodonts (after Helsen, 1995). CAI (Colour Alteration Index): *3* 120–150°C; *4* 190–245°C; *5* 245–310°C.

porosity, sedimentary rocks from Ardenne (Anceau, 1995) and other regions (Alves et al., 1995).

Control by petrology. The weakness of the diagenetic imprint resulting from the depth of burial and tectonic deformation does not imply that clay diagenesis is of minor importance in Glageon limestones. In particular the occurrence of subregular chlorite–smectite mixed-layers (i.e., corrensite) in clay mineral zones 2 and 5 is hardly compatible with syn-sedimentary formation. Corrensite is a partially-ordered transitional clay complex whose formation is not described in recent sedimentary environments (in Chamley, 1989; Weaver, 1989), and is known to form in some diagenetic environments (e.g. Desprairies and Jehanno, 1983; Thiébault et al., 1992). Typical corrensite is a trioctahedral chlorite–smectite mixed-layer that contains noticeable amounts of magnesium (Brindley and Brown, 1980). In the Glageon section, the clay mineral zone 5 (Fromelennes and upper part of Mont d'Haurs Formations) correlates with dolomite-rich lithofacies (Boulvain et al., 1994), whose depositional environment corresponded to restricted, lagoonal conditions (Fig. 3B). In addition, geochemical data show that corrensite-bearing clay fractions tend to correlate with lithologies that contain more than 3% MgO in the bulk rock (Fig. 7). These correlations suggest that corrensite is a diagenetic clay mineral which formed at Glageon under lithological control in comparatively magnesium-rich sediments. Notice that similar conditions were deduced from lithologic and isotopic investigations by Préat and Rouchy (1986), who describe pre-evaporitic conditions in several areas of the Dinant and Namur basins during Middle Devonian times, especially in the upper part of Givetian limestones which corresponds to our clay mineral zone 5. By contrast the corrensite occurring in the biostrome forming the restricted clay zone 2 (base of Trois-Fontaines Formation, Fig. 4) is characterized neither by dolomite nor by MgO, suggesting that the diagenetic mineral may not be Mg-rich and

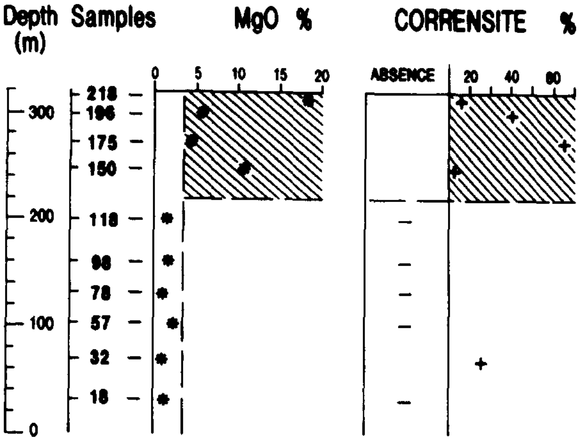

Fig. 7. Distribution of MgO in the bulk rock and of corrensite in the clay fraction from selected samples of the Glageon section. Dots indicate the wt.% MgO in the samples and crosses indicate the corresponding wt.% corrensite.

formed under another, still unknown chemical control which necessitates further, detailed petrographic and geochemical investigations.

5. Discussion of the paleoenvironmental evolution

Because the diagenetic control of clay minerals at Glageon is either of limited intensity or restricted to specific zones in uppermost Eifelian–Givetian limestones, the clay mineral stratigraphic distribution is potentially able to express successive environmental messages occurring prior to, or during Middle Devonian sedimentation. In addition, because corrensite is usually a diagenetic mineral derived from the modification of pre-existing smectite (Kisch, 1983, Velde, 1985), it seems reasonable to consider the limestones from clay mineral zones 2 and 5 as having originally contained sedimentary smectite, and to consider them in a similar way as zones 1 and 3 rocks which still include smectite in their clay fraction. Finally, because Glageon limestones are devoid of any volcanic remains (Boulvain et al., 1994), the evolution of which could potentially have induced the secondary formation of smectite, this clay mineral is considered as essentially derived from the erosion of subaerial formations exposed during Givetian time.

Smectite on land forms mainly through weathering under warm climatic conditions (Millot, 1970), as does kaolinite which is sometimes associated with smectite in the clay fractions at Glageon. Smectite and kaolinite from Glageon we therefore suppose, potentially come from the erosion of surficial soils and weathering profiles developed under a warm and more or less humid climate during Middle Devonian time in northern Europe. Such an assertion is consistent with oxygen, carbon and strontium isotope data obtained on several outcrops from the southern flank of the Dinant synclinorium by Préat and Weiss (1994) and Weiss and Préat (1994), who concluded there was a tropical humid to arid climate in the middle–late Devonian. As detrital kaolinite is usually transported over smaller distances and under stronger hydrodynamic conditions than detrital smectite (Gibbs, 1977), the relative increase of smectite amounts relative to kaolinite is tentatively attributed to more distal and quiet depositional conditions.

Mica-illite and chlorite typically constitute clay minerals formed through deep diagenetic, metamorphic or magmatic processes. Because in Glageon limestones illite and chlorite occur together with smectite and/or kaolinite which usually correspond with surface or near surface conditions (Kisch, 1983), illite and chlorite are considered to be derived from the erosion of deep diagenetic metamorphic or magmatic rocks exposed on Devonian land masses. Due to the fact that illite and chlorite at Glageon are abundant, well crystallized and ubiquitous clay minerals, it is likely that the parent-rocks responsible for their supply in the Givetian sea were widely outcropping and actively eroded in the contemporaneous river basins of northwestern Europe. The relative increase of illite and chlorite relative to smectite and kaolinite is tentatively attributed to the stronger erosion of rocky substrates compared to surficial soils, possibly because of either a tectonic rejuvenation of continentally outcropping rocks or a relative sea-level drop. Because chlorite is more sensitive to weathering than illite, the clay mineral zones enriched in chlorite can be interpreted as specifically synchronous with active tectonic events and/or sea-level lowering.

Based on the preceeding reasoning, the five clay mineral zones (Fig. 5) and thirteen carbonate microfacies identified along the Glageon section suggest the occurrence during the Givetian stage of the following environmental successions, which we place in a lithofacies and sequence stratigraphy context:

– Zone 1, latest Eifelian, early Givetian, Hanonet Formation. The occurrence in small amounts of smectite indicates open marine, rather deep water conditions favoring the accumulation of this mineral, which confirms the paleoenvironmental reconstruction deduced from carbonate microfacies investigations (Boulvain et al., 1994. See also Fig. 3A; facies 2, 3). The large abundance of illite and especially chlorite suggests active erosion on land and inferred active tectonic activity of exposed substrates. The progressive decrease of chlorite upsection could reflect the progressive decrease of tectonic movements, allowing the weathering of chlorite and the formation of soil-derived minerals (i.e., smectite and kaolinite that are present in the subsequent zones).

– Zone 2, base of Trois-Fontaines Formation. The presence of significant amounts of corrensite (25–30% of the clay fraction) in the biostromal limestones (Figs. 2 and 3A, facies 6) extending from 62.6–80 m, lacking any enrichment in magnesium (Fig. 7), is presently unexplained. One should notice the association of the diagenetic clay assemblage with fluorite, a mineral that also results from postdepositional processes, which is present within the whole Trois-Fontaines formation (Fig. 4) and whose origin probably derives from either pneumatolithic or compactional processes. The diagenetic control in the biostrome, marked by clay mineral zone 2, could be associated to a sea-level drop and related fluid migration, as suggested by vadose zone and wave-derived sedimentary structures (Boulvain et al., 1994).

– Zone 3, Trois-Fontaines Formation and base of Terres d'Haurs Formation. The presence in variable, locally high amounts of smectite (up to 45%) in the largest part of the clay mineral zone suggests conditions of settling in a quiet environment. In contrast, the occurrence of kaolinite, which is not easily transported over large distances, suggests rather deposition close to the shore, where exposed land masses constitute potential sources of the mineral. In addition the largest amounts of smectite correlate with noticeable amounts of kaolinite (Fig. 4). This apparent contradiction can be explained if the depositional environment is located close to shore in an area protected from active hydrodynamic processes by a barrier front. This is the case at Glageon during the sedimentation of the Trois-Fontaines Formation, for which we envisage the existence of a semi-lagoonal environment typical of internal shelf conditions (Fig. 3A, facies 7, 12, 13; see also Boulvain et al., 1994). Again the clay mineral characteristics fit the environmental reconstruction deduced from litho- and micro-facies data. The alternating deposition of smectite- plus kaolinite-rich clay, and of illite-rich clay (Fig. 4) could reflect alternating phases of quiet and active hydrodynamic conditions, determined by the alternation of barrier protection and wave action phases.

– Zone 4, Terres d'Haurs Formation and lower third of Mont d'Haurs Formation. The resumption of clay mineral sedimentation, rich in chlorite and illite, in a depositional environment marked by coastal conditions similar to those of zone 1, points to a major change in the clay mineral supply from exposed land-masses. The sudden disappearance of both smectite and kaolinite suggests that soil-forming processes responsible for the formation of these surficial minerals had nearly ceased. The increased amount of illite and especially of chlorite, which is more sensitive to weathering, i.e. two typically erosion-derived minerals, suggests a strong renewal of active erosional processes on land. The cause of such a renewal could be tectonic rejuvenation, a hypothesis supported by regional data on isotopic homogenization at about 373 ± 8 m.y. on magmatic activity and local mineralization (Goffette et al., 1991; Mansy and Meilliez, 1993). The tectonic instability expressed by the clay mineral associations could explain the repeated transition from open marine to more restricted environments which has been inferred from the sequence stratigraphy analysis (Fig. 2), as well as the opposition registered for Givetian time from the general eustatic curve proposed by Ross and Ross (1988). An accessory condition could have also occurred at the end of the zone 3 interval, when effacing of the barrier front during the deposition of Terre d'Haurs and first third of Mont d'Haurs Formations determined more open marine (see Fig. 3; facies 1–4) hydrodynamic conditions favoring the settling of illite and chlorite relative to smectite. Notice that according to $^{87}Sr/^{86}Sr$ data (Weiss and Préat, 1994), the sources of the Terres d'Haurs detrital supply were dominantly located to the south, i.e. towards the present-day Rocroi massif region, which was located much closer to the study area than the northern Brabant massif and could have experienced specific tectonic uplift.

– Zone 5, upper two thirds of Mont d'Haurs Formation, base of Fromelennes formation, late Givetian. The appearance of corrensite and its increase in time correlates with progressively restricted, nearshore depositional conditions, in a very coastal, chemically confined, nearly sub-aerially environment (Figs. 2, 3). Such conditions a priori favor both the synsedimentary trapping of detrital smectite in lagoonal deposits and the early diagenetic evolution of smectite into magnesian corrensite in a predolomitic environment. Notice that isotopic measurements of the contemporaneous late Givetian series from Ardenne led Weiss and Préat (1994) to suggest the occurrence of a more arid climate and pre-evaporitic conditions, both in agreement with the clay mineral distribution in zone 5.

6. Conclusions

The more than 300-m-thick Givetian platform carbonates at Glageon were deposited from about 380–374 m.y. B.P. in a small fault-controlled basin displaying outer- to inner-shelf facies. The limestones are organized in a hectometric scale in two major types of sequences, each of them evolving from open marine and relatively deep water to restricted and shallow water environments. The first type of sequence corresponds to an asymmetrical profile with successive, deep, barrier front and semi-lagoonal environments. The second type of sequence exhibits multiple deep and shallow areas that characterize a multiclinal slope which experienced an irregularly landward restriction from open marine to almost subaerially-exposed environments. 10-m- and 1-m-thick sequences occur within these large-scale sequences and could result from relative sea-level variations.

The clay mineral fraction displays diversified and variable assemblages resulting from both synsedimentary (environmental) and postsedimentary (diagenetic) influences. The moderate level of the clay mineral evolution contrasts with the strong diagenetic status of the carbonate fraction and was probably favored by early carbonate recrystallization and reduction of porosity preventing later water–rock interaction and fluid migration, as well as by the locally moderate lithostatic overburden and heating time due to early tectonic uplift. Clay diagenesis is marked by only slight effects of depth of burial and by noticeable lithological control.

The clay mineral distribution throughout the Glageon section displays a few major variations that are essentially controlled by their geographical and environmental context. Illite and chlorite represent often the dominant species and chiefly result from the erosion of nearby, actively eroded early Paleozoic substrate rocks. Smectite and kaolinite are attributed to the occurrence of a warm and rather wet climate, also suggested by biofacies as well as by isotope (Weiss and Préat, 1994) investigations. The presence of smectite-derived corrensite in latest Givetian deposits agrees with more arid climatic conditions, which also favored the smectite relative to kaolinite formation in soils. The clay mineral variations roughly correlate with major environmental successions recognized through litho/microfacies and sequence stratigraphy investigations (Fig. 3). The illite and chlorite abundance increased in hydrodynamically active environments, whereas smectite deposition was favored in deep open-sea or restricted lagoonal environments allowing differential settling processes. Corrensite is attributed to an early diagenetic evolution occurring in restricted, subevaporative environments.

The five clay mineral zones identified correlate with carbonate facies successions (Figs. 4, 5) and successively suggest: (1) a decrease of tectonic activity in open marine conditions, (2) the trapping of fine suspended material in a biostromal environment, (3) a settling in quiet back-barrier to lagoon conditions determined by the episodical instalment of a coastal barrier, (4) a tectonic rejuvenation of the exposed land masses, especially those cropping out nearby in Rocroi area, (5) and finally sedimentation in a restricted, subevaporative coastal environment.

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