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Palaeozoic clay mineral sedimentation and diagenesis in the Dinant and Avesnes Basins (Belgium, France): relationships with Variscan tectonism

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Abstract

Clay mineral investigations have been performed on more than 500 limestones and shales sampled in Lower Devonian (Emsian) to Lower Carboniferous (Namurian) outcrops in the Dinant and Avesnes Basins (Ardenne Massif, NW Europe). Clay mineral data have been placed in the palaeoenvironmental and structural histories documented by previous lithological, stratigraphical, palaeontological, diagenetic and tectonic contexts. The clay associations are dominated by illite and chlorite derived partly from the erosion of land masses surrounding the marine domain. The geothermal gradient estimated from correlation with conodont colour alteration index ranges between 40 and 70°C/km. A diachronous northwards migration of the diagenesis/metamorphism interface links to uplift caused by Late Carboniferous compressional folding and overthrusting. Associated clay minerals include smectite, locally preserved from diagenetic changes mainly by early pore closure, that reflect lagoonal or quiet offshore marine conditions. Smectite and subordinate kaolinite abundances decrease upwards during the Devonian in three successive intervals suggesting alternations of sub-arid to drier climates. The local occurrence of corrensite (ordered chlorite–smectite mixed-layer) is attributed to the moderate diagenetic transformation of pre-existing smectite

Keywords: Palaeozoic; Western Europe; carbonates; clay minerals; illite crystallinity; burial diagenesis

1. Geological setting

1.1. Geological and structural contexts

The Variscan front in Belgium and northern France is represented by Brabant para-autochthonous and Ardenne allochthonous massifs (Fig. 1). Both consist of Early Palaeozoic incompetent basement composed of siliciclastic formations with Caledonian–Acadian deformations (Michot, 1976; Meilliez et al., 1991; Mansy and Meilliez, 1993). The subsequent Palaeozoic sedimentation on the passive margin of Ardenne developed during Devonian times in small fault-controlled basins (Thorez et al., 1988; Kasimi and Préat, 1996). The Devonian sedimentary succession is characterised by increasing thickness from north to south. Land masses were located in northern Belgium up to the Carboniferous, while southern

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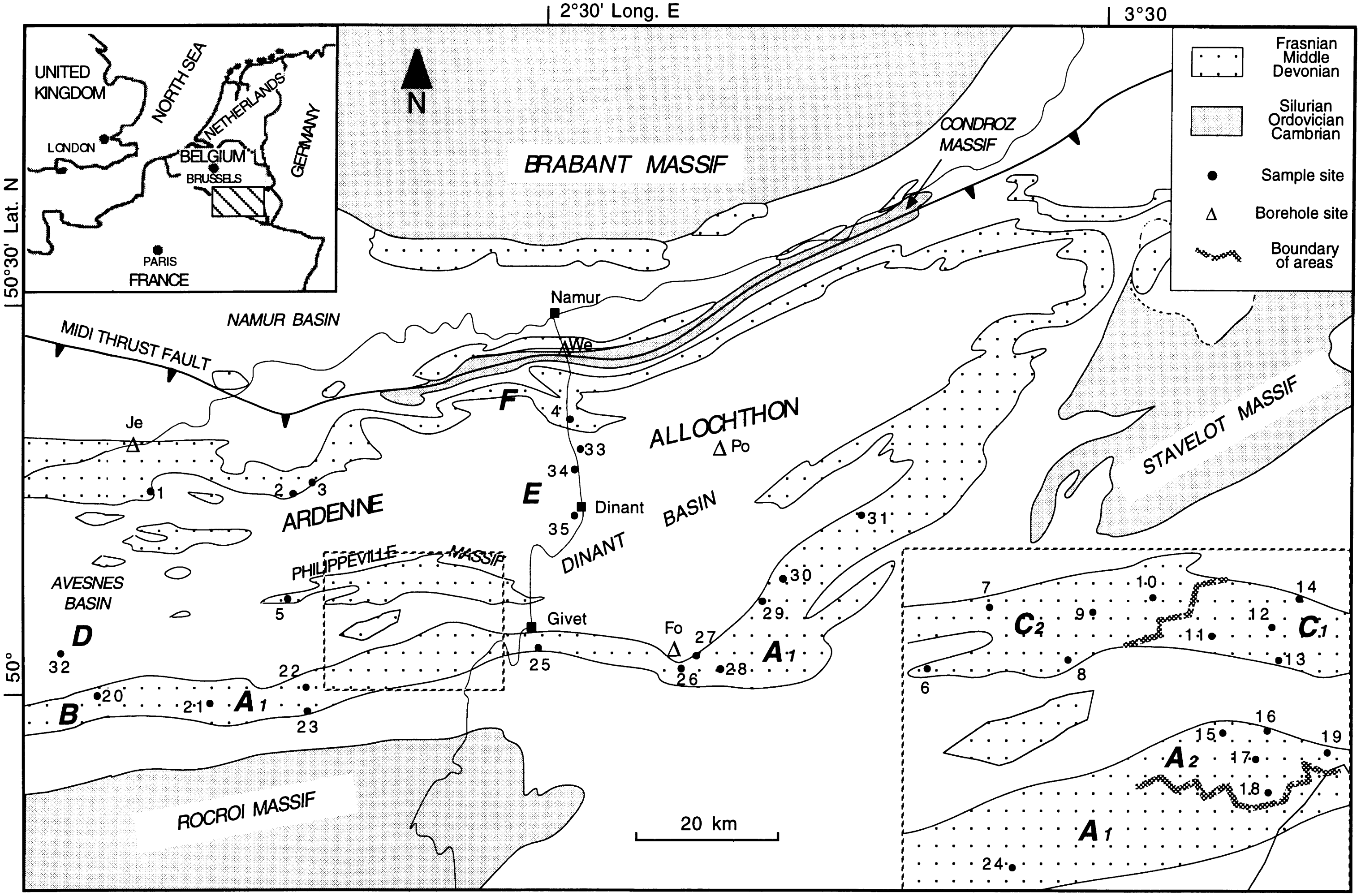


Fig. 1. Geological map of northern France and southern Belgium with location of Dinant and Avesnes Basins, and studied profiles. The names of sample sites ('O' = outcrops and 'Q' = quarries) with number are 1. Thure (Q), 2. Pry (O), 3. Gourdinne (Q), 4. Tailfer (Q), 5. Cerfontaine (Q), 6. Beauchâteau (Q), 7. N5 road (O), 8. Sautour (O), 9. Villers-le-Gambon (O), 10. Franchimont (O), 11. Marmont (O), 12. Moulin Bayot (O), 13. Hautmont (Q), 14. Soulme (O), 15. Gimnée (O), 16. Doisches (Q), 17. Vaucelle (Q), 18. Hierges (O), 19. Foisches (O), 20. Glageon (Q), 21. Villers-la-Tour (Q), 22. Haine (Q), 23. Eau Noire River (O), 24. St-Joseph (Q), 25. Flohimont (Q), 26. Wellin (Q), 27. Sourd d'Ave (O), 28. Resteigne (Q), 29. Jemelle (Q), 30. On (Q), 31. Hotton (Q), 32. Avesnes-sur-Helpe (Q), 33. Yvoir (O), 34. Anhée (O), 35. Dinant (O). The borehole names are, Fo Focant, Je Jeumont, Po Porcheresse, We Wépion. From south to north, the eight areas are as follows, A1 Southern Border of Dinant Basin (SBDB); A2 northern part of Southern Border of Dinant Basin (nSBDB); B Southern Border of Avesnes Basin (SBAB); C1 eastern part of Philippeville Massif (ePHM); C2 western part of Philippeville Massif (wPHM); D central part of Avesnes Basin (CPAB); E central part of Dinant Basin (CPDB); F Northern Border of Dinant Basin (NBDB). White areas in the Dinant, Avesnes and Namur Basins correspond to Famennian and Carboniferous.

troughs were freely connected to French basins. Hence many hiatuses occur in the Brabant area that are missing in the Ardenne (Mamet and Préat, 1996). Extensional tectonics occurred close to the Rocroi Massif, as indicated by the Middle to Late Devonian intrusion of granodiorite dykes that was followed by low-grade regional metamorphism (Goffette et al., 1991).

The thickness distribution of Devonian–Carboniferous formations shows that the depocenter from Middle Devonian (Eifelian) to Lower Carboniferous (Visean) times was located in the subsiding southern part of the Dinant Basin, forming a thick sedimentary pile of more than 3.5 km (Helsen, 1995a). The Middle and Upper Devonian exhibits unstable shelf and basin sequences (Préat and Mamet, 1989; Boulvain, 1993; Mamet and Préat, 1996) followed by a widespread Dinantian carbonate shelf (Bless et al., 1976), which later graded into paralic conditions with numerous coal seams and rare marine incursions (Kaisin, 1950). The sedimentation was abruptly interrupted mainly in the Silesian times by Variscan thrusting of the Ardenne and Dinant areas towards the Brabant Massif. The erosion of this Variscan chain was rapid, since Permian conglomerates directly cover its peneplaned roots (Mamet and Préat, 1996).

1.2. Stratigraphical context

The studied series belong to the Avesnes and Dinant Basins, including the Philippeville Massif, close to the southern border of the Dinant Basin (Fig. 1). From the top of the Lower Devonian (Emsian) to the Lower Carboniferous (base of Namurian) the series consist of an alternation of shales and limestones. The thickness of the formations ranges from dozens to hundreds of metres and sedimentation records shallow waters conditions (<150 m). The sampled formations are biostratigraphically well defined according to conodont studies (Bultynck et al., 1991). From a stratigraphical point of view the thicknesses of the Middle and Upper Devonian strata decrease drastically from 2.3 km in the south to 0.9 km in the north. Such a decrease is impossible to demonstrate for the Carboniferous strata since they are not preserved in the southern part of the Dinant Basin. The maximum average thicknesses for each geological stage are: Emsian 1000 m, Eifelian 800 m, Givetian 450 m, Frasnian 350 m, Famennian 750 m, Tournaisian 400 m and Visean 600 m (Fourmarier et al., 1954; Paproth et al., 1983; Beugnies, 1988; Bultynck et al., 1991; Adams, 1994; Helsen, 1995a; Boulvain et al., 1999), and between 1 and 4.5 km for the Upper Carboniferous, deduced from Conodont Alteration Index (CAI) studies (Helsen, 1995a).

1.3. Purpose

The objectives of this paper are to: (1) document the relationship between sedimentary facies and clay mineral distribution to identify the respective palaeoenvironmental and diagenetic influences; (2) compare the illite crystallinity (IC) evolution through the stratigraphic series and between distinct sub-basins; (3) determine the geothermal gradients of these Variscan basins; and (4) integrate the data from sedimentary successions and IC evolution in a new Variscan tectonic model.

2. Material and methods

We have sampled the Devonian and Carboniferous series of the Dinant and Avesnes Basins as well as the Devonian series of the Philippeville Massif. The sedimentological context is well known (Préat and Mamet, 1989; Boulvain, 1993; Kasimi and Préat, 1996; Chamley et al., 1997) and constitutes the basis of this work. 509 samples were collected (473 limestones and 36 shales) from outcrops of each Palaeozoic formation. All selected carbonate samples were petrographically studied and the main characteristics of the environment were based on microfacies studies (see Préat and Mamet, 1989; Préat and Kasimi, 1995).

Clay mineral associations have been investigated using X-ray diffraction analysis of oriented mounts. Deflocculation of clays was performed by successive washing with distilled water after decarbonatation of the crushed rock with 0.2 N HCl. The clay fraction (<2 μm particles) was separated by sedimentation and centrifugation (Brindley and Brown, 1980; Holtzapffel, 1985). X-ray diagrams were obtained using a Philips PW 1730 diffractometer with CuKα radiations and Ni filter. A voltage of 40 KV and a current of 25 mA were utilized. Three X-ray diagrams were performed after air-drying, ethylene-glycol solvation and heating at 490°C for 2 h. The goniometer scanned from 2.5 to 28.5° 2θ for air-dried and glycol-solvated conditions and from 2.5 to 14.5° 2θ for heating conditions. The identification of clay minerals was made according to the position of the (00l) series of basal reflections on the three X-ray diagrams (Brindley and Brown, 1980; Reynolds, 1980; Moore and Reynolds, 1989). Semi-quantitative estimations were based on the intensity (peak height *H*) and on the area (compared with standards to determine the coefficient *C*) of the main diffraction peak of each clay mineral, with a calculation: the abundance of a given mineral (a) equals to $(H_a C_a / \sum H_i C_i)100\%$ (after Holtzapffel, 1985). Analytical uncertainties in the abundance of chlorite, illite, smectite, kaolinite and random mixed-layers are estimated at ±3% for abundances above 20%, ±2% for abundances between 20 and 5%, and ±1% for abundances below 5%. Illite, smectite and chlorite crystallinity indices (IC, SC and CC) that correspond respectively to the width at half peak height of 001, 001 and 002 peaks after glycolation have been measured. The mean errors equivalent to the combined effects of measurement error, machine variation and intra-sample variation (Robinson et al., 1990) range from 3.0 to 7.3%, comparable to the error interval estimated by Kisch, (1990: from 3.3 to 8.3%) and by Warr and Rice (1994: from 2.9 to 7.4%). Four standard samples have been investigated enabling the calibration of the illite crystallinity data using the crystallinity index standard (CIS) of Warr and Rice (1994) with good correlation ($R^2 = 0.982$) (Robion et al., 1999; Averbuch, Lille University, pers. com).

3. Results

3.1. Clay stratigraphical distribution

The clay fraction of the studied Palaeozoic rocks (representing about 1–5% of the bulk rock) is characterised by an unexpected diversity of minerals and strong variations in their abundance. The clay minerals include mica-illite (0–100%), chlorite (0–62%), smectite (0–64%), kaolinite (0–48%), corrensite (0–80%) and random mixed-layers (0–33%). The average abundances for all samples are the following: mica-illite 70%, chlorite 16%, smectite 7%, corrensite 4%, kaolinite 2% and random mixed-layers 1%. The clay assemblages show important differences depending on the stratigraphical level, the geographical location and the lithology (shales or limestones). Fig. 2 shows the clay distributions in the Middle Devonian formations on the southern border of the Dinant Basin. Illite is by far the most abundant clay mineral within the series, with an average between 60 and 90% of the total clay content. The illite abundance presents a weak regular gradient from lower values at the base (Couvin Formation) to higher values at the top (Fromelennes Formation). However this trend, balanced by decreasing proportions of chlorites cannot be generalized. The Eifelian and the Givetian are characterised by decreasing proportions of smectite and kaolinite from base to top.

3.2. Crystallinity index

3.2.1. General values

Analysis of the IC values allows recognition of eight areas within the studied region. From south to north they are (see Fig. 1):

A1: southern border of Dinant Basin (SBDB),
A2: northern part of southern border of Dinant Basin (nSBDB),
B: southern border of Avesnes Basin (SBAB),
C1: eastern part of Philippeville Massif (ePHM),
C2: western part of Philippeville Massif (wPHM),
D: central part of Avesnes Basin (CPAB),
E: central part of Dinant Basin (CPDB),
F: northern border of Dinant Basin (NBDB).

These eight subdivisions (from A1 to F) are consistent with the regional geology: they correspond to the main structural parts of the Dinant Synclinorium (parts A1, B, D, E, and F) and are based on facies distribution for the remaining areas (A2, C1 and C2).

IC values were measured and an IC zonation was used following Kübler (1967) and Yang and Hesse (1991).

Zones	IC (°$\Delta 2\theta$)
Higher part of high diagenetic	0.62–1.00
Highest grade diagenetic	0.42–0.62
Lower anchimetamorphic	0.33–0.42
Higher anchimetamorphic	0.25–0.33

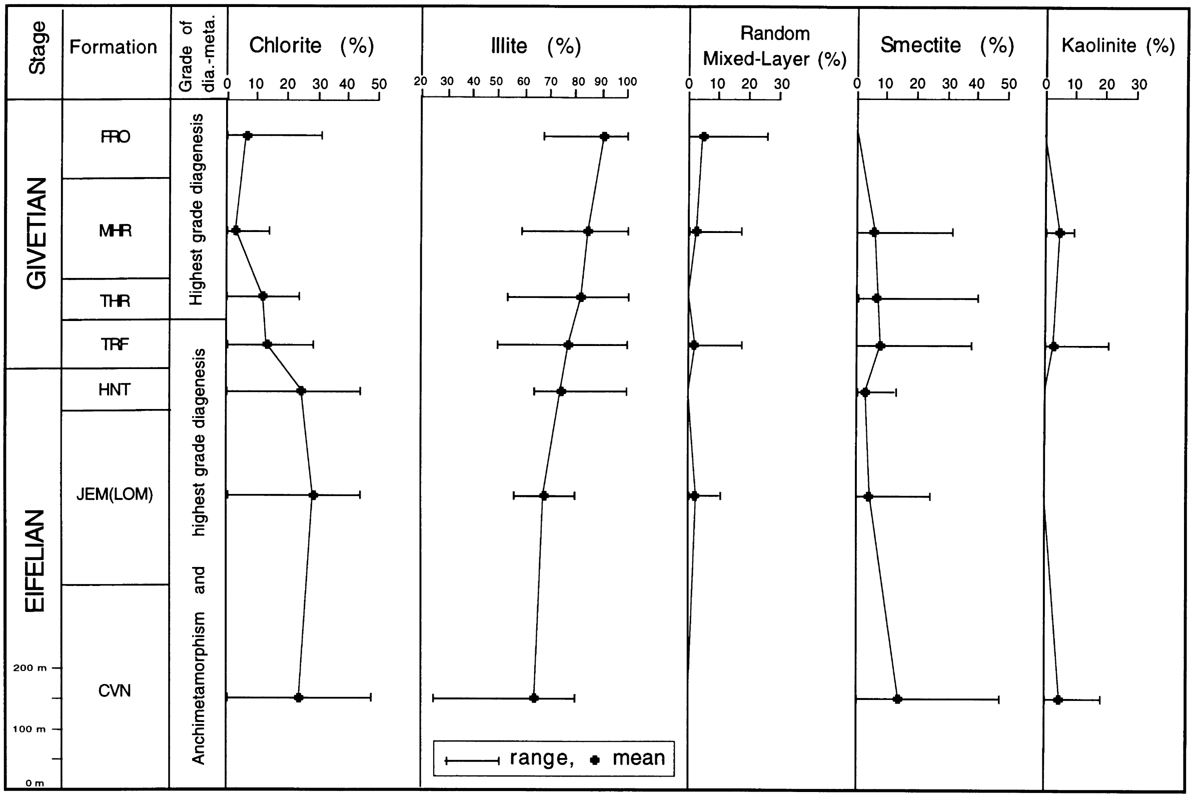


Fig. 2. Formations and clay mineral distributions in the Middle Devonian limestones of the southern border of the Dinant Basin. CVN = Couvin Formation, JEM (LOM) = Jemelle (Lomme) Formation, HNT = Hanonet Formation, TRF = Trois-Fontaines Formation, THR = Terres d'Haurs Formation, MHR = Mont d'Haurs Formation and FRO = Fromelennes Formation (after Bultynck et al., 1991).

The IC values ranging from 0.86 to 0.27° $\Delta 2\theta$ do not depend on the lithology. The highest values (4% of the total samples) indicate the higher part of the high diagenetic zone and the lowest ones (3%) belong to the higher anchizone. Most of the values belong to the highest grade diagenetic zone (63%) and to the lower anchimetamorphic zone (30%).

3.2.2. Stratigraphic distribution and location of the diagenesis/anchimetamorphism interface

The vertical variation of IC in most of the eight studied areas shows a quite normal linear trend without internal offsets (Figs. 3–5). Fig. 3 shows the IC distribution, based on 120 samples from 11 sites, as a function of the stratigraphic levels in the southern border of the Dinant Basin. Most calculated averages are reliable according to their histogram, standard deviation and the sample number of each formation. The distribution of IC values from Lower Devonian to Upper Givetian is regular from the anchimetamorphic zone (shales and quartzose siltstones of the Lower Devonian and Lower Eifelian) to the diagenetic zone (limestones and shaly limestones of the Upper Givetian). The position of the diagenesis/anchimetamorphism interface occurs at the transition between the Eau Noire Formation and the Couvin Formation (Fig. 3). A similar gradient is also present in the Avesnes Basin; for example, in the southern border (Fig. 4a) the IC distribution is equally from the anchimetamorphic zone (shaly limestones of the Upper Eifelian) to the diagenetic zone (shaly dolomites of the Upper Givetian) with interface occurring between the Mont d'Haurs Formation and the Fromelennes Formation.

The vertical profile of IC in the central part of the Dinant Basin does not fit the general linear trend

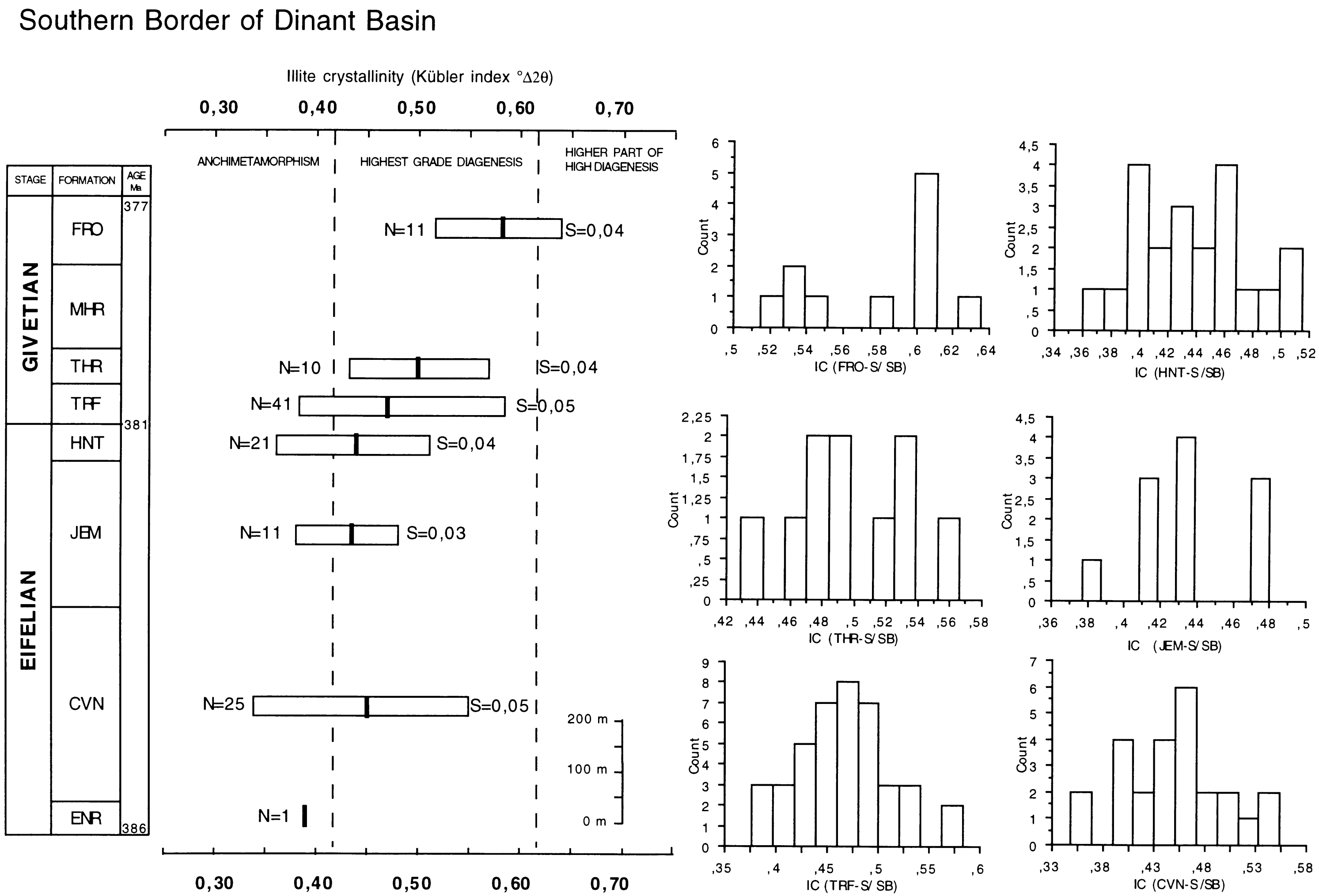


Fig. 3. Illite crystallinity distribution in the Middle Devonian of the southern border of Dinant Basin. Formations are the same as Fig. 2 (ENR = Eau Noire Formation). Legend: N = count number, S = standard deviation, black vertical bar is the average. Most of the data for each formation presents a normal distribution (see histograms).

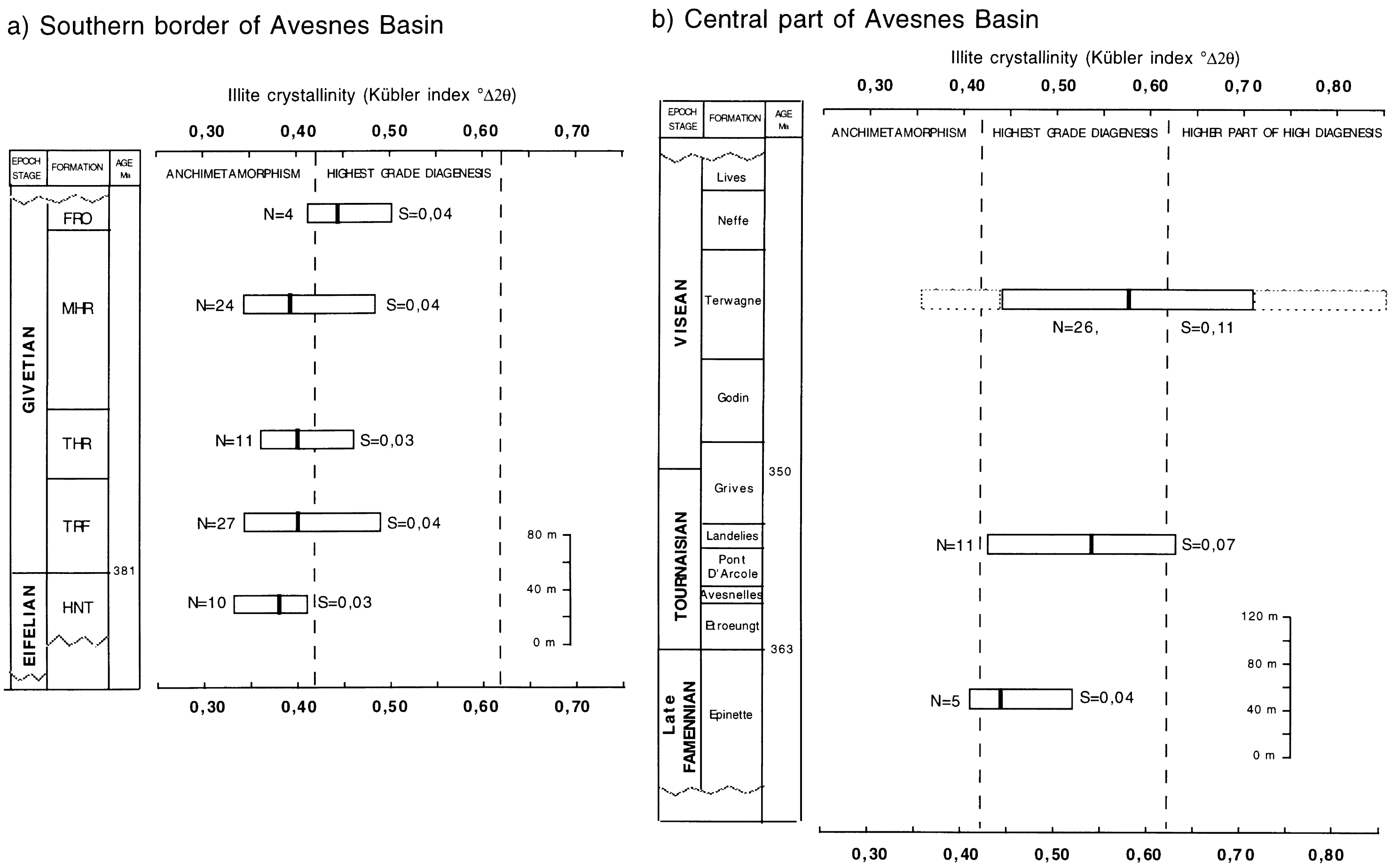


Fig. 4. Illite crystallinity distribution in the southern border (a) and the central part (b) of the Avesnes Basin. Middle Devonian Formations are the same as Fig. 2. Lower Carboniferous Formations after Mansy et al. (1989). Legend: N = count number, S = standard deviation, black vertical bar is the average of the values for each formation (Southern Border) or stage (central part).

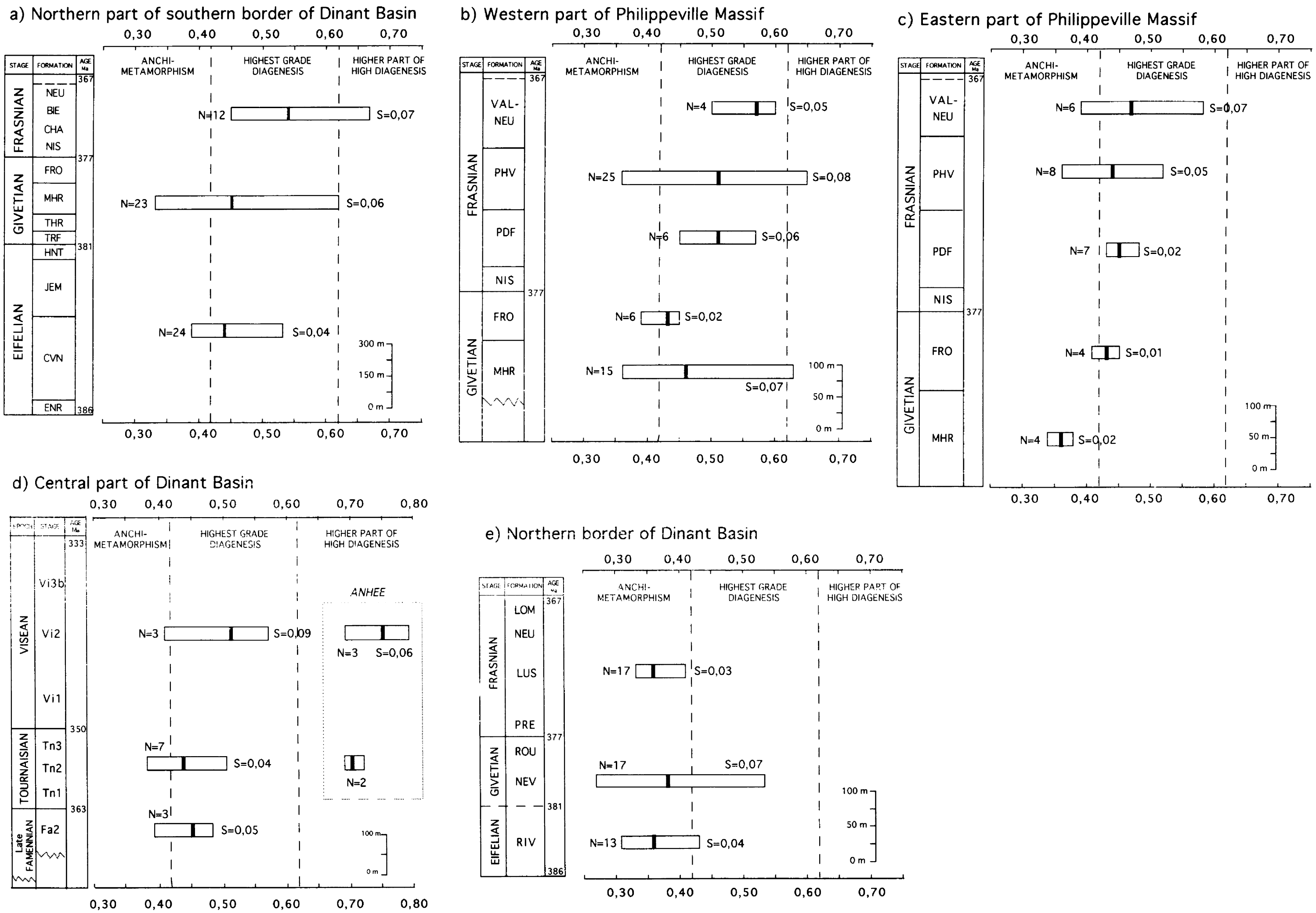


Fig. 5. Illite crystallinity distribution in the northern part of Southern Border (a), central part (d) and Northern Border (e) of the Dinant Basin, western (b) and eastern parts (c) of the Philippeville Massif. All the Formations as Figs. 2 and 3. For the central part of Dinant Basin, Fa2 = Upper Famennian, Tn1,2,3 = Lower, Middle and Upper Tournaisian, V1,2,3 = Lower, Middle and Upper Visean. For the northern border of Dinant Basin, RIV = Rivière Formation and NEV = Nèvremont Formation after Lacroix (1974). Legend: N = count number, S = standard deviation, black vertical bar is the average.

without some internal offset. A diagenetic gap is present at least in the Lower Carboniferous around the Anhée area located a few kilometres north of the Dinant locality (Figs. 1 and 5d). Such a deviation from a normal linear gradient is also confirmed by the IC analyses of Dandois (1981).

IC distribution in each domain shows a normal burial trend, except in the northern part of the Dinant Basin (Fig. 5e), where no gradient is observed since all the IC values remain in the anchizone, which is probably due to incomplete stratigraphical sequences. But it can be reasonably deduced that this interface position lies in the Famennian series according to the general sedimentary burial trend and the pre-Famennian IC distribution within the anchizone near the diagenetic zone (Fig. 5e).

The IC stratigraphic distribution of the eight areas reveals an important pattern, besides the ubiquitous IC gradient, which is the shifting of diagenesis/anchimetamorphism interface northwards to younger strata in both studied basins (Fig. 6).

4. Discussion

4.1. Origin and significance of some clay minerals

4.1.1. Smectite

The abundance of chlorite and illite suggests a diagenetic influence. Similar clay assemblages were previously described in the Upper Devonian of the Boulonnais area (northern France) and also interpreted in terms of burial (Lefrançois et al., 1993).

The large amount of data available on carbonate microfacies (Préat and Boulvain, 1987,1988; Préat and Mamet, 1989; Boulvain, 1990; Préat and Kasimi, 1995) provides a good basis for a hydrodynamic analysis of the sedimentary environments. The general comparison between the distribution of smectite and facies analysis in carbonate platform and mixed ramp systems shows that about 94% of total samples with smectite occur in low to moderate hydrodynamic conditions. A similar association of smectite with low energy environments is reported in the Upper Devonian, Lower Carboniferous and Mesozoic carbonate platforms (Darsac, 1983; Adatte and Rumley, 1984; Trentesaux, 1989; Huyghe and Trentesaux, 1992). Correlation of smectite with hydrodynamic conditions supports a sedimentary or depositional genesis for this mineral. The occurrence of smectite in Palaeozoic series of Ardenne was expected as this mineral was previously noticed in Devonian limestones of the Glageon quarry (Avesnes Basin, Chamley et al., 1997). At Glageon, smectite is considered as being dominantly a mineralogical relict of terrigenous material, since no lithological, sedimentological or geochemical argument can be found to support any hydrothermal or volcanic influence. For the same reason this interpretation is proposed for explaining the occurrence of smectite in the sections and boreholes investigated in the Dinant Basin.

Illitization of smectites during diagenesis needs a source of K and Al (Hower et al., 1976; Frey, 1987). Chemical analyses of our samples by XRF show that limestones have little K and Al (respectively less than 0.5 and 1.3%) and that shales contain much more K (5.8%) and Al (22%) (Boulvain, 1981; Préat et al., 1983; Dejonghe, 1985). As reported by Chamley et al. (1997) in the Glageon limestones, the early diagenetic cementation of the carbonate rocks reduced the porosity and prevented later fluid–rock interaction. All our samples show also an early cementation as reported by Weis and Préat (1994). These two reasons, lack of K and Al, and occlusion of the porous system by early cementation, probably allowed smectite to be preserved during the highest grade diagenesis and the anchimetamorphism. This situation also exists in other carbonate regions where smectite may even persist into the epizone (Frey, 1987).

Preserved smectites likely suffered re-crystallization during diagenesis and anchimetamorphism. Smectite crystallinity and abundance show that crystallinity in the lower anchizone (with mean of $0.47° \Delta 2\theta$) is higher than in the highest grade diagenetic zone (with mean of $0.57° \Delta 2\theta$) within the five studied regions (Table 1). Illitization is almost absent since the abundance of smectite is larger in the anchizone, i.e. in the more buried zones (Table 1).

4.1.2. Corrensite

Variable amounts of subregular chlorite–smectite mixed-layers of the corrensite group occur in the Givetian, the uppermost Tournaisian and the Visean of the Avesnes region (Avesnes-sur-Helpe and Glageon, Chamley et al., 1997).

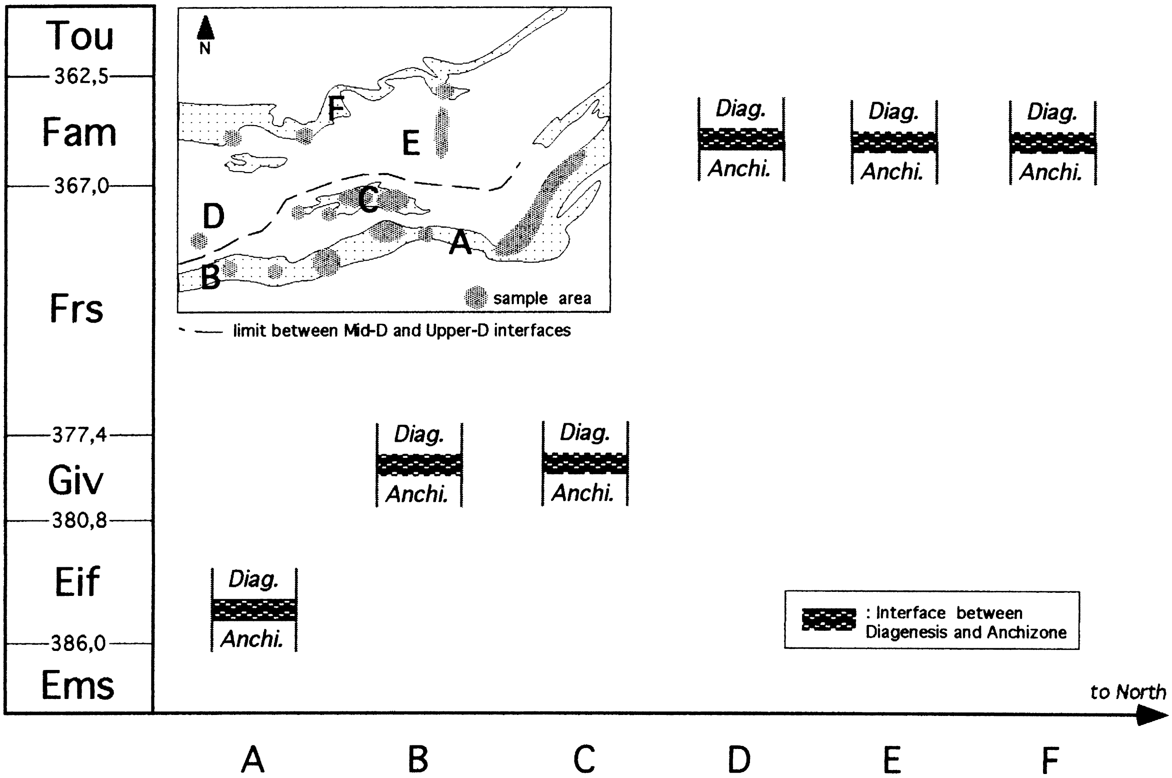


Fig. 6. Stratigraphical and geographical migration of the diagenesis/anchizone interface in the Avesnes and Dinant Basins. Ems = Emsian, Eif = Eifelian, Giv = Givetian, Frs = Frasnian, Fam = Famennian and Tou = Tournaisian. Time scale from Harland et al. (1989). The names of areas within the studied region are, A Southern Border of Dinant Basin (SBDB); B Southern Border of Avesnes Basin (SBAB); C Philippeville Massif (PHM); D central part of Avesnes Basin (CPAB); E central part of Dinant Basin (CPDB); F Northern Border of Dinant Basin (NBDB). The diagenesis/anchizone interfaces of both basins display northern younger migration.

Corrensite corresponds to depositional environments marked by quiet hydrodynamic conditions. This is the case both at Glageon within penecontemporaneous dolomites from restricted, low energy environments (Fromelennes Formation, Upper Givetian; see also Chamley et al., 1997), and at Avesnes-sur-Helpe (Bocahut quarry) within secondary dolomites from open marine, slowly settling conditions (Grives

Table 1
Distribution of chlorite and smectite crystallinities (° $\Delta 2\theta$) with smectite abundance in the Avesnes and Dinant Basins including the Philippeville Massif. Numbers in brackets are the count numbers. CC = chlorite crystallinity, SC = smectite crystallinity, S% = smectite abundance, A.B. = Avesnes Basin, D.B. = Dinant Basin, Ph.M. = Philippeville Massif. The crystallinities of chlorite and smectite are not calibrated

Zone		A.B.	Southern border of D.B.	Western part Ph.M.	Eastern part of Ph.M.	Central part of D.B.	Northern border of D.B.
Highest grade diagenetic	CC	0.26 ($N = 12$)	0.29 ($N = 39$)	0.33 ($N = 8$)	0.31 ($N = 10$)	0.26 ($N = 5$)	
	SC	0.57 ($N = 9$)	0.51 ($N = 7$)	0.56 ($N = 7$)	0.62 ($N = 10$)		
	S%	17	18	37	38		
Lower anchizone	CC	0.22 ($N = 15$)					0.23 ($N = 7$)
	SC	0.48 ($N = 17$)	0.43 ($N = 7$)		0.50 ($N = 4$)		0.49 ($N = 5$)
	S%	15	30		52		19

Formation, Tournaisian-Visean; see Trentesaux, 1989).

Corrensite, the formation of which is unknown in recent sedimentary environments (e.g. Chamley, 1989; Weaver, 1989), is considered to result from a moderate diagenetic evolution of detrital minerals. The similarity observed between smectite and corrensite inferred environmental conditions suggests that corrensite derived from the diagenetic evolution of smectite that had initially settled in lagoonal or moderately energetic offshore areas (Bouquillon et al., 1985). Notice that corrensite has also been interpreted as resulting from the diagenetic evolution of terrigenous clay minerals, in the lacustrine carbonate mudrocks of the Devonian of the Orcadian basin in Scotland, where the burial depth was estimated to correspond to a temperature of about 100°C (Hillier, 1993).

4.2. Palaeoclimatical significance

The geographic distribution of smectite in 10 Middle and Upper Devonian formations shows a clear pattern. Three intervals occur and are distributed as follows (Fig. 7): interval 1 from Middle to Late Eifelian, interval 2 from Early to Late Givetian and interval 3 from Early to Late Frasnian. Within each interval the mean smectite content is systematically higher at the base (between 10 and 15%) and decreases towards the top ($<5\%$). As smectite is considered as dominantly inherited from subaerial erosion, its changing distribution within each time interval could result from climatic variations on exposed land masses surrounding the Devonian basin. Smectite on land forms mainly through weathering under warm conditions (Millot, 1970). In the sections investigated it is occasionally associated with kaolinite (see for example Fig. 2), a clay species that also develops dominantly through chemical alteration under a warm climate. Such a clay mineral association constitutes an argument supporting a palaeoclimatic interpretation.

The soil-derived origin of smectite and kaolinite from the Dinant Basin (where our data are the most complete) is supported by additional arguments of both lithological and geochemical nature. Thirteen palaeosoils have been recognized by Yans et al. (1997) in the Middle Devonian (Eifelian/Givetian transitional beds) at Remouchamps (northeastern border of the Dinant Basin). The presence of abundant smectites associated with other pedogenetic structures (glaebules, rhizolites, pisolites, …) suggests a warm climate with alternating wet and dry seasons. This assertion is consistent with Sr isotope analyses (Weis and Préat, 1994), with stromatolitic lamination types (Boulvain and Préat, 1986; Préat and Boulvain, 1986) and primary dolomite and evaporite distribution during Givetian times (Préat and Rouchy, 1986; Préat and Mamet, 1989). Finally the upwards decrease of the proportion of smectite, a mineral that usually characterises sub-arid climatic conditions, could correspond to the progressive transition to a drier climate in the course of Givetian times.

4.3. Comparison between IC and other heat-related indicators

4.3.1. CC and SC versus IC gradient

Many researches relating to clay mineral crystallinity indicate that the growths of chlorite and illite coexist during the evolution of diagenesis and very low-grade metamorphism (e.g. Warr, 1996; Arkai et al., 1996), although their growth rates are different under anchizonal and epizonal conditions (Merriman et al., 1995). In our study, chlorite and smectite crystallinities (respectively CC and SC), which were investigated in 96 and 66 measurable samples were statistically compared with IC zones. Table 1 shows CC and SC values in the different areas as a function of the diagenetic and anchimetamorphic zones. The values shows increasing recrystallization from highest grade diagenetic zone (0.26–0.33° $\Delta 2\theta$ for CC and 0.51–0.62° $\Delta 2\theta$ for SC) to low anchizone (0.22–0.23° $\Delta 2\theta$ for CC and 0.43–0.50° $\Delta 2\theta$ for SC). This increase of CC and SC with burial supports the widespread existence of the IC gradients previously reported.

4.3.2. Illite crystallinity versus conodont alteration index (CAI)

Helsen (1992, 1995a,b) has published more than 500 conodont colour alteration indices from Belgium. The error is less than half an index by comparison with the CAI standards within a Palaeozoic population containing more than 50 specimens (Helsen, 1995b). We compared them with our IC values in

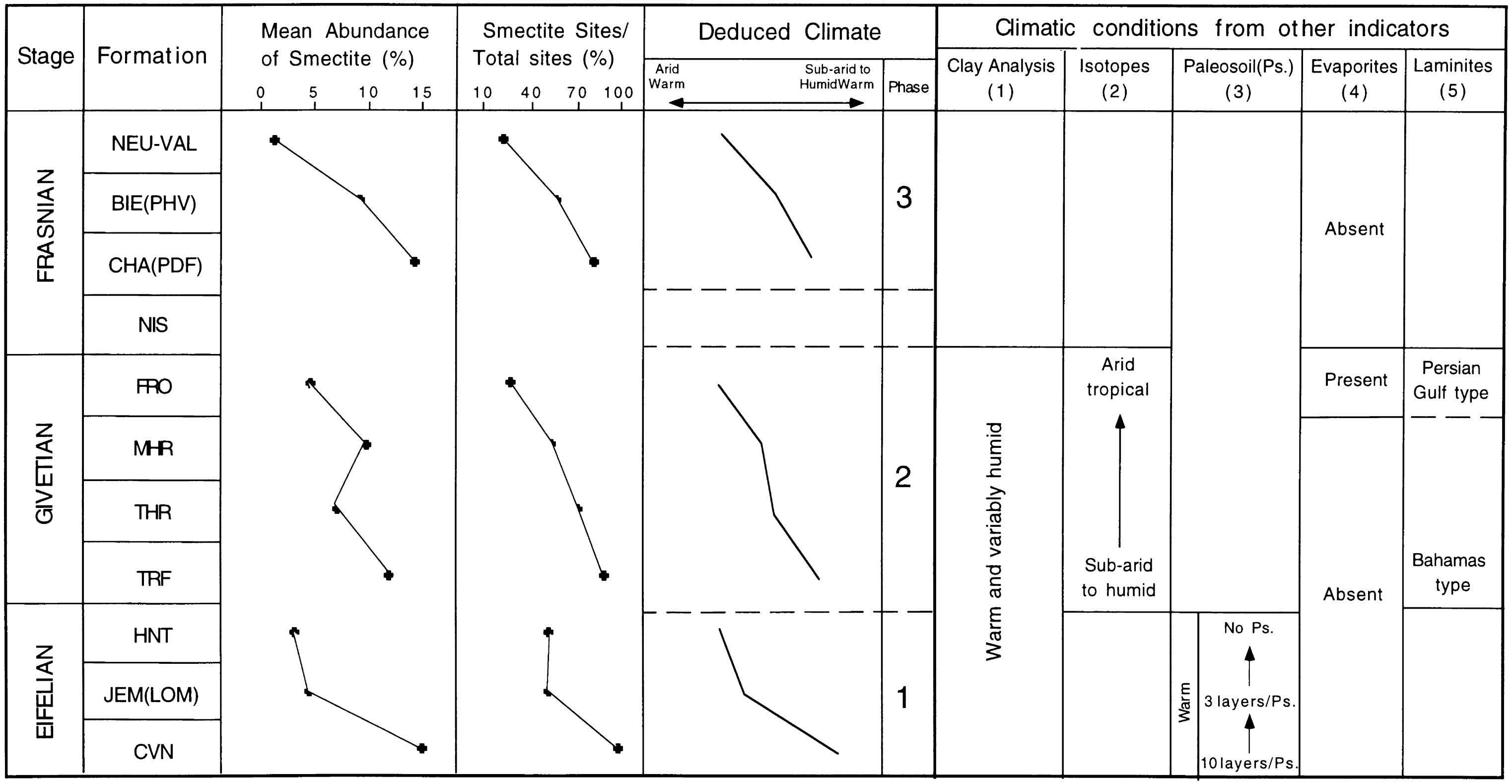


Fig. 7. Stratigraphical and geographical distribution of smectite and deduced palaeoclimate in the Middle and Upper Devonian of the studied region. Eifelian and Givetian Formations are the same as Fig. 2. For Frasnian Formations: NIS = Nismes Formation, CHA (PDF) = Châlon (Pont de la Folle) Formation, BIE (PHV) = Bieumont (Philippeville) Formation and NEU (VAL) = Neuville (Valisettes) Formation (after Boulvain et al., 1993; Boulvain et al., 1999). Columns (1) after Chamley et al. (1997), (2) Weis and Préat (1994), (3) Yans et al. (1997), (4) Préat and Rouchy (1986) and (5) Préat and Boulvain (1986).

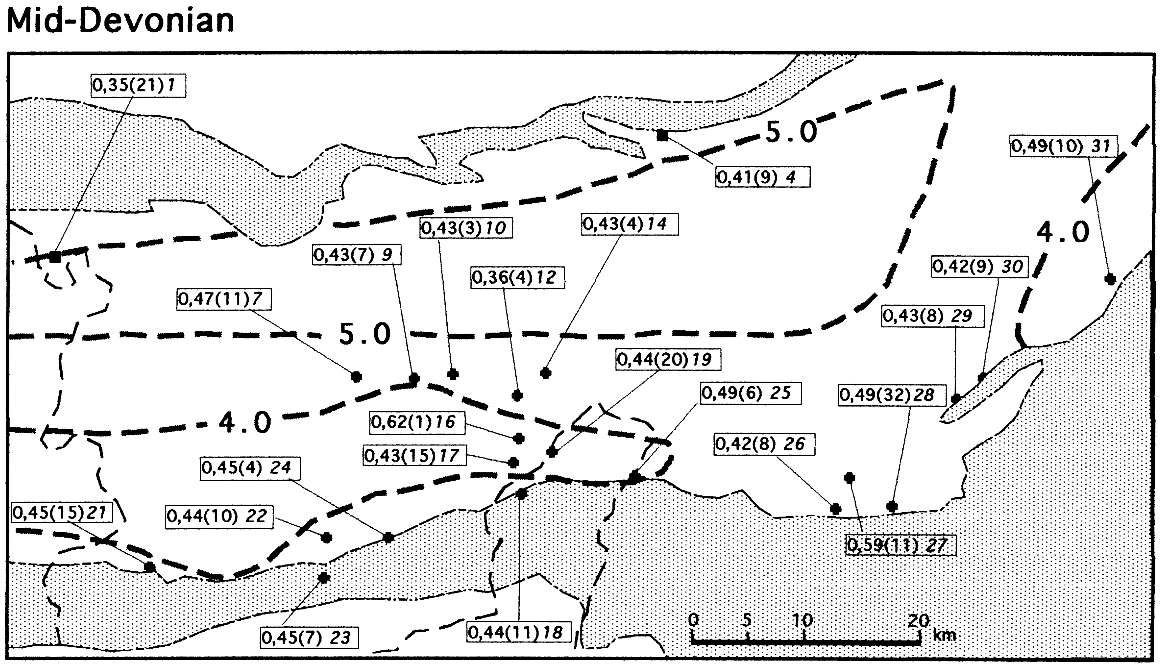


Fig. 8. Distribution of illite crystallinity on Conodont Alteration Index map of Helsen (1992) for the Middle Devonian of Dinant Basin. The numbers on isograds represent the CAI values. Within each rectangle, first number is the mean of IC (CIS), second number is sample count, the last one represents the sample site as follows. 1. Thure, 4. Tailfer, 7. N5 road, 9. Villers-le-Gambon, 10. Franchimont, 12. Moulin Bayot, 14. Soulme, 16. Doisches, 17. Vaucelle, 18. Hierges, 19. Foisches, 21. Villers-la-Tour, 22. Haine, 23. Eau Noire, 24. St-Joseph, 25. Flohimont, 26. Wellin, 27. Sourd d'Ave, 28. Resteigne, 29. Jemelle, 30. On, 31. Hotton.

the Dinant Basin by superimposing IC maps to the CAI's maps of Helsen (Fig. 8). This work encompasses only the Mid-Devonian and Frasnian intervals where both IC and CAI's values are abundant enough. The 311 IC values that are present on the superimposed maps can be divided into three groups according to their location in the 5.0, 4.5 and 4.0–3.5 CAI isograds or zones (Fig. 8). The IC values were analysed with boxplot in Statview 4.0 program in order to assess the average and to identify the representative range for each IC group (Moore and McCabe, 1989 pp. 35–38; Haycock et al., 1992 pp. 387–390). The result shows that the ranges and the means of IC groups of both stages are almost the same in the identical CAI zones and that the illite crystallinity increases from 4.0–3.5 to 5.0 CAI zones (Table 2). This table also gives the correlation between CAI zones and the IC zones. Some CAI/IC values dealing with different Palaeozoic stratigraphies and tectonic settings in southern (Garcia-Lopez et al., 1997) and northern Europe (Kisch, 1987), and in North America (Kisch, 1987) compare well with those of this study.

The correlation of IC values with CAI zones and with temperatures (Table 2) is very useful for estimating geothermal gradients (see Section 4.5.1) that cannot be otherwise derived from IC values. In fact the data of the literature concerning the temperature range for anchizone display a broad dispersion: this temperature ranges between 280 and 360°C (Weaver and Boekstra, 1984), between 200/250 and 300°C (Kisch, 1987), and between 175 and 320°C (Warr, 1996). The problem is similar with oxygen isotopes (Primmer, 1985) or fluid inclusions (Niedermayr et al., 1994). The thermal alteration of conodonts gives more precise temperature ranges (Epstein et al., 1977; Rejebian et al., 1987).

4.4. IC profiles of some boreholes

The collected IC data of four boreholes (see Fig. 1)

Table 2
Comparison between the IC and CAI values in the Dinant Basin including the Philippeville Massif. In columns of IC values, numbers in brackets are the count numbers, other numbers are means and ranges. See the text for details

Equivalent temperature (burial time: 30–80 MA Helsen 1992, 1995a)	CAI value	IC value (° Δ2θ)		IC zone (° Δ2θ)	Equivalent temperature (Kisch, 1987)
		Mid-D	Frasnian		
310–300°C	5.0	$\overline{0.35}$ (21) 0.31–0.41	$\overline{0.36}$ (17) 0.33–0.41	Lower anchizone 0.33–0.42	
245–240°C	4.5	$\overline{0.43}$ (38) 0.35–0.51	$\overline{0.43}$ (12) 0.36–0.50	Transition (anchi/diagenetic) ± 0.42	200–250°C (at boundary between anchi-and diagenetic zones)
195(190)–150(145)°C	4.0–3.5	$\overline{0.46}$ (167) 0.40–0.53	$\overline{0.51}$ (56) 0.43–0.60	Highest grade diagenetic 0.42–0.62	

in the studied region provide a chance to observe deep IC distribution compared with their surface equivalents. The Focant borehole shows a strongly imbricated 3 km-thick Frasnian series (Fig. 9). The same series are about 350 m thick in the southern border of Dinant Basin and in the Philippeville Massif. The IC profile of the Focant borehole (Goudalier, 1998) is composed of two parts: the lower part (‘part B’) displays for the same Boussu-en-Fagne Member (from member 7 to member 1) both clear CAI and IC gradients recording a re-heating from tectonic overloading. The upper part (‘part A’) retains similar IC values as those reported in the Frasnian of the southern border of Dinant Basin (Fig. 5a).

The IC data of other boreholes mainly display redistribution features of remained sedimentary burial IC depending on the form of tectonic stacking. The IC values of the Jeumont borehole (Bouquillon et al., 1985) show that the Devonian series, which overlie the Carboniferous strata due to thrusting along the Midi Fault, are more mature than the latter. A symmetrical repetition of IC caused by the syncline structure appears in the Devonian of the Wépion borehole (Dandois, 1985). The sedimentary burial gradient due to surviving of a normal stratigraphical sequence is retained in the Lower Devonian at depth interval from 2000 to near 5000 m in the Porcheresse borehole (Dandois, 1985). But this profile also shows an inverted diagenesis between the Middle and Lower Devonian (i.e. Middle D. is more mature than lower D.); in this case no associated faulting is reported and the explanation remains open.

Analysis of all IC data (outcrops and boreholes) permits a general understanding of IC genesis in the studied region. Most of the surface and near-surface IC keep the sedimentary burial origin based on the wide existence of the IC stratigraphic gradients, while the IC in some tectonically deepened strata (especially young ones) can be re-improved due to larger tectonic loading. part B of the Focant profile rarely shows the typical characteristics of the latter.

4.5. Basin evolution

4.5.1. Estimation of the geothermal gradient

In order to compare the IC gradients between the different regions, the cumulative thicknesses and IC mean values are used to draw representative gradient lines. According to regression plots (Statview 4.0) their correlation coefficients are high, averaging 0.88 and ranging from 0.76 to 0.997. When placed in an IC/thickness diagram (Fig. 10a) the gradient lines can be grouped in two main domains with a small third one. The first group (SBDB, nSBDB and CPDB) shows slopes in the range of 0.010 to 0.015° Δ2θ/100 m. The second group (wPHM, CPAB and SBAB) displays slopes between 0.030 and 0.040° Δ2θ/100 m. The third group (only ePHM) has an intermediate slope (0.025° Δ2θ/100 m).

Analysis of the groups shows two main geothermal

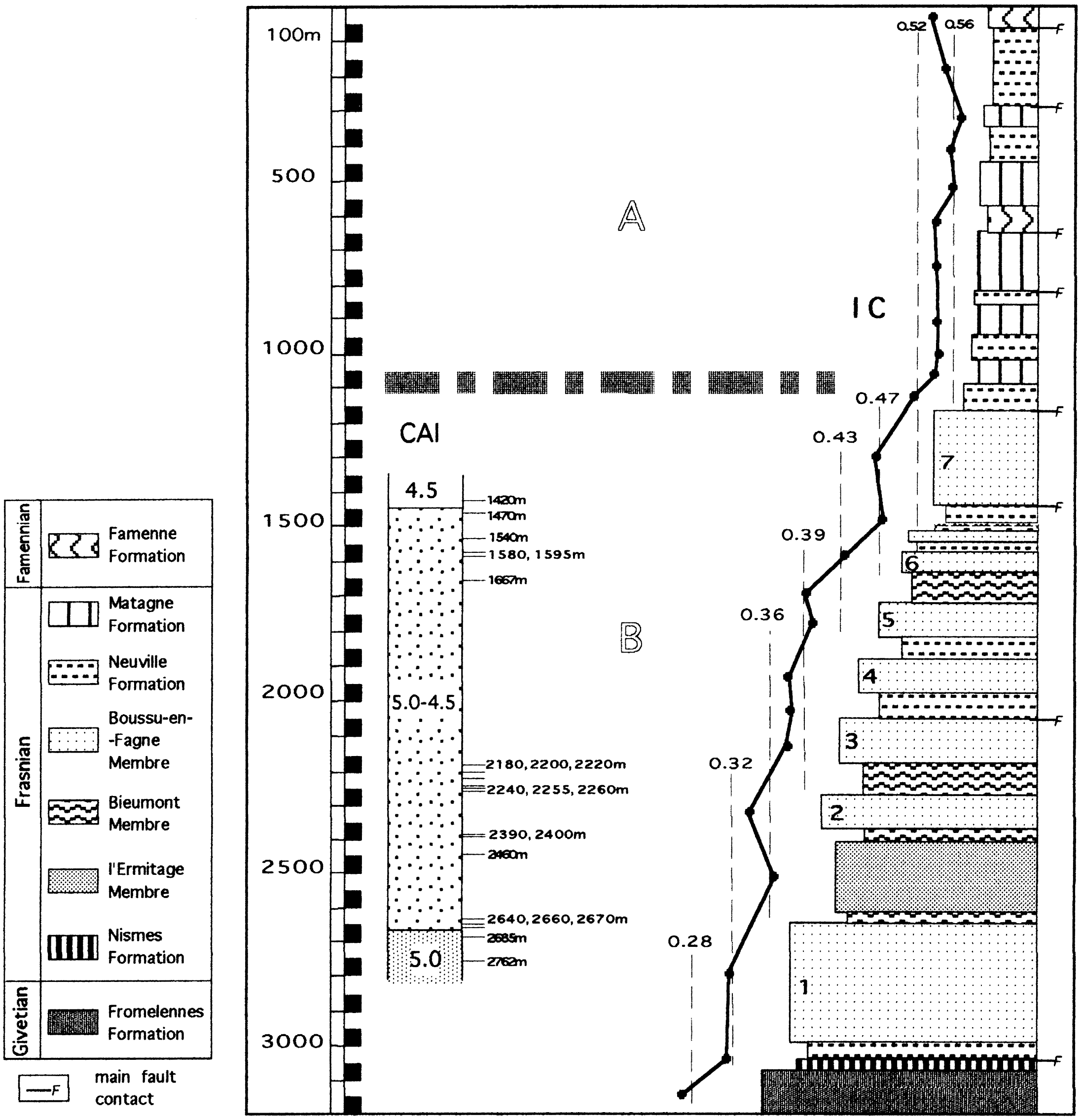
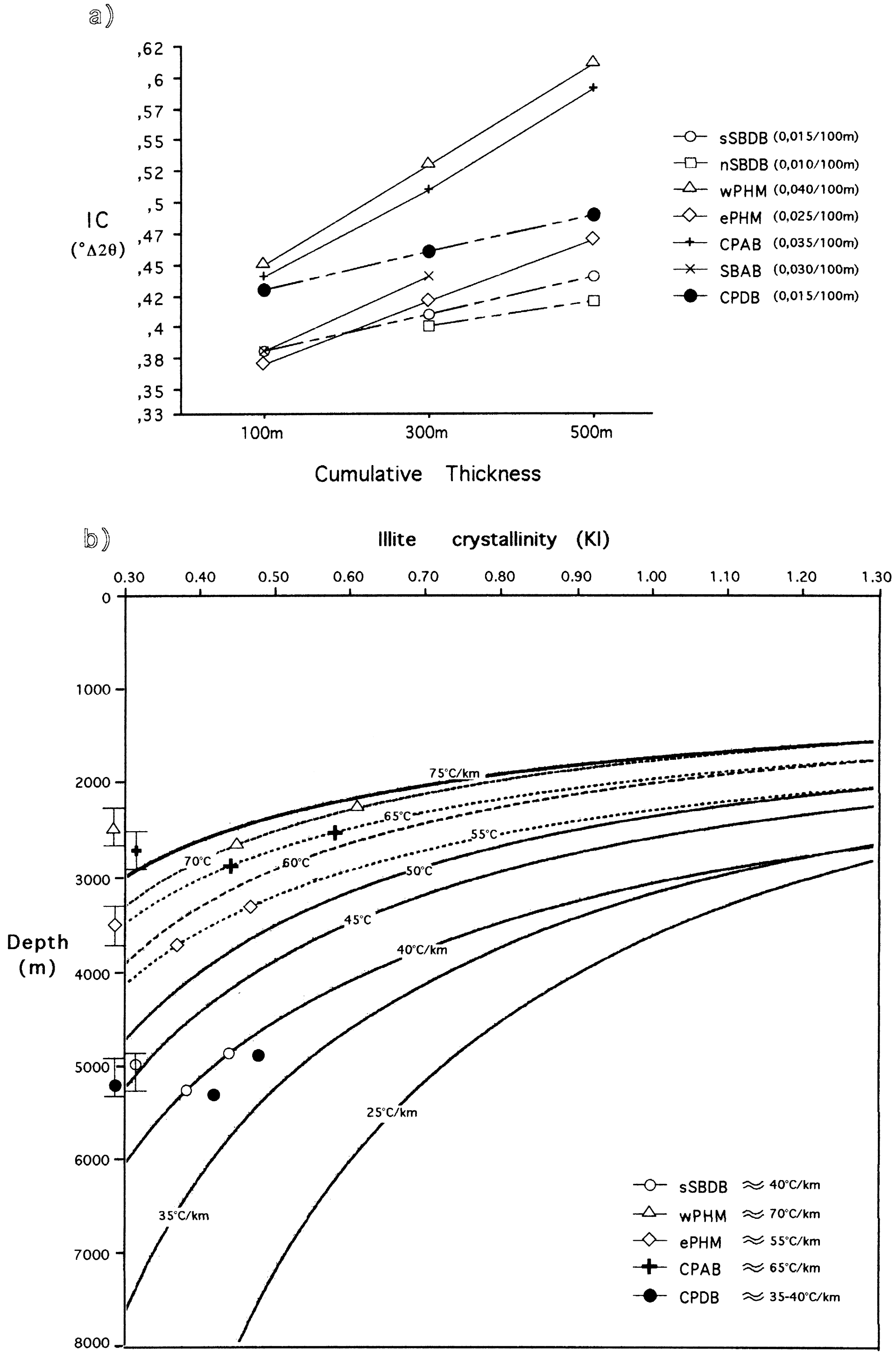


Fig. 9. IC and CAI distributions in the Frasnian formations of the Focant borehole. The upper part (part A) shows IC values in the range of those of the southern border of the Dinant Basin (see Fig. 5a). The lower part (part B) displays both IC and CAI gradients (especially IC evolution in the seven tectonic-repeated layers of Boussu-en-Fagne Member from numbers 7 to 1) recording a re-heating event from tectonic loading. IC data are from Goudalier (1998), CAI data are from Helsen (1995b).

gradients: one for the Dinant Basin (southern and central parts) and another for the Avesnes Basin (southern and central parts) and the wPHM. The ePHM with a slope intermediate between these two groups could belong to the first one based on its stratigraphical context (Boulvain et al., 1993). The second group presents a higher geothermal gradient since its slope is more pronounced, and both groups have comparable and limited IC interval (0.36–0.60° $\Delta 2\theta$, see Figs. 3–5).

The IC gradients of the two preceding groups together with temperature data from CAI (Helsen,


a)
IC
(°Δ2θ)
,62
,6
,57
,55
,52
,5
,47
,45
,42
,4
,38
,35
,33
100m
300m
500m
Cumulative Thickness
sSBDB (0,015/100m)
nSBDB (0,010/100m)
wPHM (0,040/100m)
ePHM (0,025/100m)
CPAB (0,035/100m)
SBAB (0,030/100m)
CPDB (0,015/100m)
b)
Illite crystallinity (KI)
0.30
0.40
0.50
0.60
0.70
0.80
0.90
1.00
1.10
1.20
1.30
Depth
(m)
0
1000
2000
3000
4000
5000
6000
7000
8000
75°C/km
65°C
55°C
70°C
60°C
50°C
45°C
40°C/km
35°C/km
25°C/km
sSBDB ≈ 40°C/km
wPHM ≈ 70°C/km
ePHM ≈ 55°C/km
CPAB ≈ 65°C/km
CPDB ≈ 35-40°C/km

1995a) allow an estimation of the geothermal gradients during the sedimentary burial times. According to the correspondence recorded between the CAI 4–3.5 isograd and the highest grade diagenetic zone (Table 2), the 45°C temperature interval of the CAI 4–3.5 isograd can be approximately considered as the temperature interval of the highest grade diagenetic zone where the IC interval is about 0.20° $\Delta 2\theta$. The calculations using IC and thickness intervals permit to establish the IC slopes under some assumed geothermal gradients. After comparing these slopes with the measured IC slopes of the seven regions (Fig. 10a), the preliminary geothermal gradients are estimated. They are then compared with those resulting from another method (Fig. 10b) using an independent set of IC vs depth curves from computer modelling based on data of the Gulf Coast shales (Merriman, pers. com.) to estimate the geothermal gradients. The comparison shows they are essentially consistent except for some slight differences appearing above 60°C/km. The geothermal gradients with new constraint are estimated as follows: about 40°C/km for the Dinant Basin and ePHM, 70°C/km or so for the Avesnes Basin and wPHM.

These gradients are in the range of those indicated by numerous thermal maturation studies in the European and North African fold belts and foredeep basins, which are characterised by gradients varying between 40 and 70°C/km (in Helsen, 1995a). Moreover, in the Campine Basin, located immediately north of the Brabant Basin in Belgium, a gradient of 47°C/km was calculated from fluid inclusions by Muchez et al. (1991) in the same Palaeozoic sequences. The higher gradient could explain the strong dolomitization (with very coarse xenotopic textures) of the series in the Avesnes Basin and in the wPHM or the importance of Pb–Zn–(Ba–F) mineralizations in the wPHM (Préat et al., 1983). Mineralizations of this type tend to be associated with high heat flow (Cauet and Weis, 1983) in sedimentary basins in back-arc settings (Smellie et al., 1996). Such a setting may have characterised the region described here, as suggested on a broader scale by Ziegler (1990).

4.5.2. Structural implications

IC depth profiles that result from continuous sedimentation in a subsiding basin are characteristic of many coeval sub-basins in this area (Chamley, 1989). These profiles indicate that burial diagenesis played an important role in the different parts of the basin as indicated by the illite crystallinity data, CAI analyses and isograd patterns from Belgium (Dandois, 1981; Helsen, 1995b). In general the maturation of Palaeozoic strata is considered to be pre-tectonic (Oncken, 1984). This simplified evolution has to be reviewed, however, in the light of our results, which are mainly based on the basinwide migration of the diagenesis/anchimetamorphism interface through time.

Assuming a surface temperature of 20°C and an interface temperature of 220°C (Kisch, 1987), the calculated sedimentary loadings at the interfaces are about 5 km with geothermal gradient of 40°C/km (for the Dinant Basin) and 3.3 km with a mean geothermal gradient of 60°C/km (for the Philippeville Massif). These values can be equally obtained on Merriman's IC/depth curves (Fig. 10b). The known Devono-Dinantian thickness (except for a CAI deduction for the Dinantian on the southern border and the Philippeville Massif) above the interface is 3450 m for the southern border, 1850 m for the Philippeville Massif, 1300 m for the central part and 1250 m for the northern border (Fourmarier et al., 1954; Paproth et al., 1983; Beugnies, 1988; Bultynck et al., 1991; Adams, 1994; Helsen, 1995a; Boulvain et al., 1999). As there are no significant post-Variscan sediments in the studied region (Ziegler, 1990;

Fig. 10. (10a) comparison of IC gradients among the seven areas. Two major IC gradient groups can be recognized according to their slopes. Legend: The right numbers are the slope values in ° $\Delta 2\theta$/m. SBDB = Southern Border of Dinant Basin, nSBDB = northern part of Southern Border of Dinant Basin, wPHM = western part of Philippeville Massif, ePHM = eastern part of Philippeville Massif, CPDB = central part of Dinant Basin, CPAB = central part of Avesnes Basin, SBAB = Southern Border of Avesnes Basin. (10b) IC versus depth curves with geothermal gradients from computer modelling of the Gulf Coast shales (USA). According to the IC slope of each studied areas, the thickness interval of 400 m and its corresponding IC range (from Fig.10a) are placed on this diagram and checked for each curve. The good ones are indicated on this figure. The result shows that the geothermal gradients deduced from this figure and from CAI temperature are essentially consistent except slight difference appearing above 60°C/km. After Merriman, unpublished data form the Gulf Coast (USA).

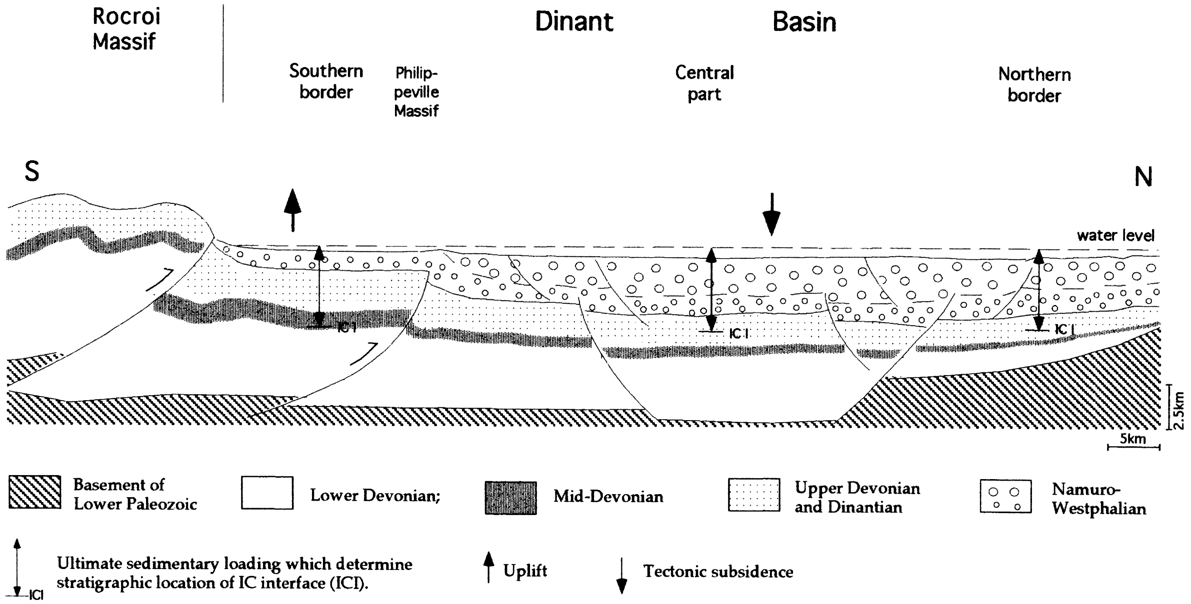


Fig. 11. The sequence of overthrusting-induced uplifts causes the differential sedimentary wedges in the Dinant Basin during the Namuro-Westphalian times. Uplift first took place in the southern border of the Dinant Basin (including Philippeville Massif), shutting down sedimentation in early Namurian times. Meanwhile the central and northern parts continued to subside and accepted large amount of sediments. Finally the northwards migration of the Variscan uplift caused the shutdown of sedimentation in the central and northern parts in late Westphalian times. The northern younger IC interfaces (diagenesis/anchimetamorphism) preserve a record of this tectonic evolution.

Meilliez et al., 1991), the deduced thickness of the Namurian–Westphalian series is, about 1.5 km for the southern border of the basin and the Philippeville Massif, and about 3.5 km for the central part and the northern border.

The Silesian strata of the studied region formed in a foreland basin characterised by compression (Bless et al., 1989; Ziegler, 1990; Meilliez et al., 1991). The general evolutionary pattern of the foreland basin can be considered as a process of continual progress towards the foreland of thrust-induced uplift in the hanging wall and frontal basinal subsidence in the footwall (Einsele, 1992). In this way, the later uplifted central and northern parts would have received a thicker fill than earlier uplifted southern parts (including the Philippeville Massif).

This uplift sequence is strongly supported by the data from previous isotopic dating. The K–Ar ages of illites (<2 μm) from Devonian rocks in the Dinant Basin are respectively 326 ± 7 Ma in the southern border (Piqué et al., 1984) equivalent to the early Namurian (Ziegler, 1990, Encl. 45), and 297 ± 7 Ma in the central part (Piqué et al., 1984) equivalent to the late Westphalian (Ziegler, 1990, Encl. 45). These illites are believed to form at their maximum depth of sedimentary burial based on geological setting of the basin and ubiquitous IC gradients; the isotopic ages above should correspond to the timing of uplift.

From the evidence presented above the early extensional phase in the Dinant Basin was followed by compression, inversion and thrust fault induced uplift. This uplift began on the southern border of the basin and on the Philippeville Massif (Fig. 11), causing a shutdown in sedimentation in early Namurian times. Meanwhile the central part and the northern border of the basin continued to subside and receive a thick Silesian fill as a consequence of thrust-sheet and sediment-load induced flexural downbending of the foreland. The foreland migration of the Variscan uplift caused the shutdown of the sedimentation in the central part and the northern border of the basin in late Westphalian times. The northern younger IC interfaces (diagenesis/anchimetamorphism) preserve a record of this tectonic evolution.

In this context the reported IC values can be related to a pre-orogenic Variscan burial metamorphism for most of the Devonian and Carboniferous rocks on the southern border, and to a synorogenic Variscan metamorphism for the Middle Devonian to Upper Carboniferous fill of the central and northern parts of the Dinant Basin (Fielitz and Mansy, 1999).

5. Conclusions

The regional study of Palaeozoic clay minerals in northern France and southern Belgium has outlined a series of IC evolutions and palaeotectonic elements relating to sedimentary history within the eight recognized areas. The most important of these elements is the diachronous migration of the diagenesis/anchimetamorphism interface from southern to northern parts. This result was unexpected since Devonian strata generally thicken by ratios of 1:3 to 1:5 from the northerly Brabant Massif towards the south. In fact, this interface migration is directly related to a notable difference of Silesian sedimentary thickness between South and North, and records a successively northwards shifting of uplift caused by the Variscan thrusting–folding. This intepretation is consistent with a general evolution of a foreland basin and the previous isotopic ages of the regional burial metamorphism. The interface, based on ubiquitous IC gradient, is therefore for the first time used here to link intra-basinal tectonic–sedimentary evolution concerning the sedimentary shutdown that first took place in early Namurian times in the southern border, and then in late Westphalian times in the central part and in the northern border.

IC data and thickness distributions in combination with colour alteration of conodonts allow estimation of the geothermal gradients. The estimation was further compared with that based on the IC vs depth curves from Gulf Coast shales (USA). Two main geothermal gradients are recognized: the first one in the Dinant Basin (southern and central parts) with an average of 40°C/km and the second one in the Avesnes Basin (southern and central parts) and in the wPHM with an average of about 70°C/km.

Analysis of all IC data (outcrops and boreholes) suggests that most of the surface and near-surface IC kept the sedimentary burial origin based on the wide existence of their stratigraphic gradients, while the IC in some tectonically deepened strata (especially young ones) could be re-improved due to subsequent larger tectonic loading from the Variscan thrust.

Smectite is locally preserved from diagenetic changes mainly by early pore closure of the carbonate rocks. The local occurrence of corrensite (ordered chlorite–smectite mixed-layer) is attributed to the diagenetic transformation of pre-existing smectite. Analyses of the clay mineral assemblages also indicate that several palaeoclimatological variations occurred during the Devonian times from warm subarid to arid and pre-evaporitic climates.

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