

# Trends in Early Cretaceous clay mineralogy in NW Europe

By

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With 12 text figures and 1 table

## ABSTRACT

Distinct trends occur in the clay mineralogy of fine-grained sediments from the Portlandian to Aptian in NW Europe. In many instances, these trends reflect variations in the weathering conditions that existed in the source-areas and can be used to help reconstruct the source-area environment. The following general trend in clay mineralogy can be recognised in most areas of NW Europe. From the Portlandian, through the Berriasian and into the Valanginian, kaolinite increases in abundance whilst the mixed-layer minerals decrease and change in composition. This reflects a transition in the source-areas from low relief beneath a hot semi-arid climate, to-

ward relatively high (re-emphasised relief) beneath a warm humid-temperate climate. The soils and saprolites changed from alkaline and poorly-leached ('pedocals') to acid and well-leached ('podzols'). From the Hauterivian, through the Barremian and into the Aptian, kaolinite gradually declines whilst the mixed-layer minerals increase and their composition again changes. This trend reflects gradual submergence of the land-masses beneath shallow shelf-seas which effectively lowered source-area relief, so making leaching less severe, and also perhaps in part a climatic amelioration.

## KURZFASSUNG

In den feinkörnigen Sedimenten des Portland bis Apt in NW Europa lassen sich Unterschiede in der Zusammensetzung der Tonminerale feststellen. Solche Entwicklungstendenzen spiegeln öfters die Verwitterungsbedingungen in den Ursprungsgebieten wider und helfen bei der Rekonstruktion ihrer Umweltsbedingungen. Folgende Veränderungen und allgemeine Entwicklungstendenzen in der Tonmineralogie können in den meisten Gebieten NW Europas aufgezeigt werden: Vom Portland, während des Berrias bis in das Valangin wächst der Gehalt an Kaolinit, während die Wechschelschicht-Minerale ("mixedlayer minerals") abnehmen und ihre Zusammensetzung ändern. Die zeigt den Übergang von einem flachen Relief unter heißem semiariden Klima zu einem

relativ ausgeprägten Relief unter warmem, feuchttemperiertem Klima an. Die Böden und Verwitterungsdecken wechselten von alkalisch und schwach ausgelaugt ("pedocals") zu sauer und stark ausgelaugt ("podzols"). Vom Hauterive, durch das Barreme bis in das Apt nimmt der Kaolinit allmählich ab, während die Wechschelschicht-Minerale zunehmen und auch in der Zusammensetzung sich verändern. Diese Entwicklung zeigt stufenweises untertauchen vom Land unter einem flachen Schelfmeer an, dies hatte eine Verflachung des Reliefs im Ursprungsgebiet zur Folge; so wird die Auslaugung abgeschwächt. Vielleicht steht hiermit auch eine Klima-Verbesserung im Zusammenhang.

## INTRODUCTION

Clay minerals that have recently been transported and deposited on continents, on continent margins and in the ocean basins frequently carry characteristics inherited from their source-areas. Often, there is a correlation between the source-area weathering environment and clay mineral assemblages

(WEIR et al. 1975; FEUILLET & FLEISCHER 1980; SCOTT 1975). The weathering environment (or type of leaching that occurs) is the interactive result of the climate (particularly temperature and rainfall), the topography, the types of vegetation and organisms, the parent rock composition and length of time involved. This paper examines the origins of clay minerals in the Early Cretaceous sediments of NW Europe with particular emphasis on the weathering environment in source areas. Due to a coincidence of features, the weathering environment

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can be deduced for sediments supplied to many areas e. g. Lower Saxony, the Weald and Wessex.

Samples of clays and fine-grained silts from each of the areas studied (Fig. 1) have been analysed using X-ray diffraction techniques which allow a precise identification and semi-quantification of the clay minerals present (SLADEN, 1980). Analysis of hundreds of samples coupled with the use

of trends in relative clay mineral abundances prevents overinterpretation of data which are known to be only semi-quantitative. Careful, reliable analyses are required because variations in the assemblages, which are dominated by kaolinite, illite and mixed-layer minerals, are often quite subtle. The techniques used were designed to recognise these variations.

TRANSPORT, DEPOSITION, DIAGENESIS AND RECENT WEATHERING

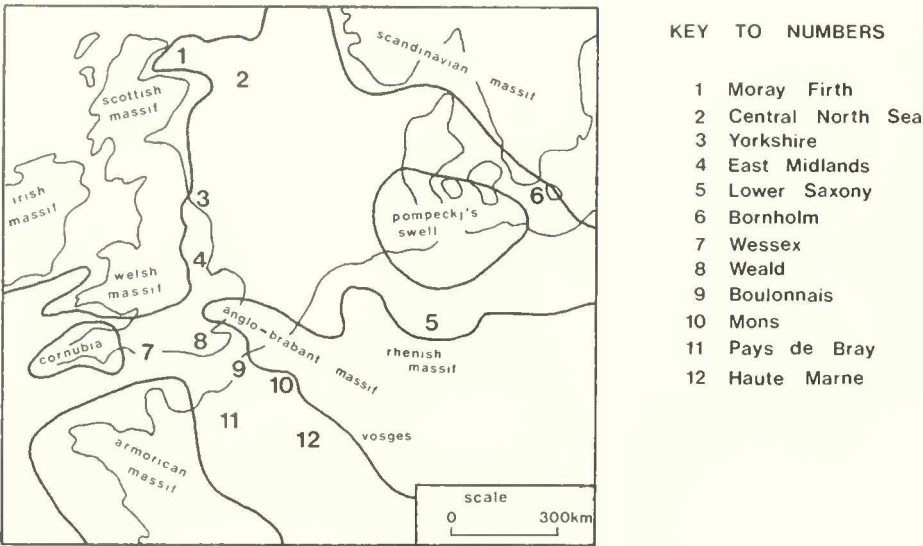


FIG.1. SIMPLIFIED PALAEOGEOGRAPHY OF NW EUROPE IN THE EARLY CRETACEOUS AND AREAS WHERE THE SEDIMENTS HAVE BEEN STUDIED

DIAGENETIC GRADE	PARAMETERS INDICATING DIAGENETIC GRADE				YORKSHIRE				BORNHOLM				WEALD				BOULONNAIS				MONS			
	illite crystallinity index	illite sharpness ratio	% 2M illite	polymorph maximum burial temperature																				
	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D
immature	>20	<1.5	<20	<70	31	11	11	40	25	13	8	45	30	11	10	65	23	14	14	35	29	12	0	32
mature	16-20	1.5-2.0	20-50	70-140																				
super mature	<16	>2.0	>50	>140																				

TABLE 1. DIAGENETIC GRADE OF EARLY CRETACEOUS FINE-GRAINED SEDIMENTS FOR VARIOUS AREAS OF OUTCROP IN NW EUROPE



Unveiling the origins of clay minerals in clay sediments must proceed in a methodical order because they are extremely sensitive to environmental changes. The first feature to consider is the effect of recent weathering. Borehole samples and fresh outcrop samples from similar horizons usually show similar assemblages even of very sensitive mixed-layer minerals and this indicates that there is little alteration occurring near the surface (full discussion in SLADEN, 1980).

The next effect to consider is diagenesis. Various parameters of the illites and mixed-layer illite-smectites, together with organic maturation levels, indicate an immature diagenetic grade and thus little alteration due to burial (Table 1). Calculating a maximum burial depth also indicates that conditions were not sufficient to alter the clay minerals (even with a high geothermal gradient). (Some alteration has occurred in the more deeply buried areas, particularly some parts of the North Sea, but these samples are not considered here). Early diagenesis has also left the clay minerals generally unaltered. Siderite ironstones formed prior to significant burial and hydromorphic gleys developed sphaeorsiderite and iron oxide

mottles but these horizons show little change in the clay minerals (SLADEN, 1980 & in press.). Rare volcanogenic horizons were transformed into smectite-rich bentonites (O'B KNOX & FLETCHER, 1978).

The various clay mineral assemblages that are found all occur in fresh water, brackish and marine sediments and this indicates that halmrolysis did not markedly affect them during transport and deposition. Also, size-grading of the clay minerals during transport was very limited. This was because there were only small differences in the particle sizes of the kaolinite, illite and mixed-layer minerals and also, the distance of transport was often very short (less than 500 km in many instances).

The discussion above indicates that, in many samples, the clay minerals survived transportation, deposition and diagenesis without significant modification to their structures or relative abundances. In these samples, the clay mineralogy must closely reflect the products of the weathering environment in their source areas.

## ASSEMBLAGES AND TRENDS CREATED BY SOURCE-AREA WEATHERING

In this, and the following sections, only the clay mineralogy of samples unaffected by transport, deposition and diagenesis is considered. This mineralogy is taken to be the product of source-area weathering environments.

### GENERAL FEATURES

Upward, from the late Portlandian through to the early Aptian, a distinct trend in clay mineral abundances occurs. From the late Portlandian into the Berriasian, kaolinite increases and mixed-layer minerals decrease. Kaolinite reaches a maximum during the late Berriasian and Valanginian and then from the Hauterivian onwards, kaolinite gradually declines and mixed-layer minerals increase. The decline in kaolinite content continues at least into the early Aptian. In areas where the sequence is fairly complete, the above trend is well-developed (Figs. 2–5). In other areas, where the sequences are incomplete parts of the trend can still be recognised (Figs. 6–11).

At the same time that the abundance of clay minerals was changing, so too was the nature of many clay minerals, particularly the mixed-layers. Also, the various clay minerals making up an assemblage often varied slightly from area to area across NW Europe and so this creates an 'identify' to many areas. These are described in more detail below.

### SOUTH OF THE ANGLO-BRABANT MASSIF

The increase in kaolinite and decrease in mixed-layers during the late Portlandian and Berriasian is very marked. The more complete sequences in the Wessex-Weald Basin show a rapid increase in kaolinite around the Jurassic-Cretaceous

boundary (Fig. 2). As kaolinite increases through the Berriasian, the mixed-layers decrease and change in character. In sediments where kaolinite is scarce, the mixed-layer mineral is typically a 10–14 Å illite-smectite with fairly random interstratification. Where kaolinite is common or abundant, the main mixed-layer mineral is Al-hydroxy vermiculite with some illite interlayering. These associations are also observed in the ensuing Cretaceous. Thus, as kaolinite decreases through the Hauterivian, Barremian and early Aptian, illite-smectites become more common and Al-hydroxy vermiculites gradually disappear. The general trend in clay minerals occasionally shows slight variations. Notably in the Weald, there are temporary reductions in kaolinite (and the reappearance of illite-smectites) at those times in the Valanginian dominated by argillaceous sediments.

Apart from illite-smectite and Al-hydroxy vermiculite, other mixed-layer minerals are generally scarce. Occasionally, there is vermiculite-smectite and chlorite-vermiculite. In the Mons Basin, there are significant amounts of Al-rich smectite (beidellite) and also chlorite-smectite, whilst in Wessex, there are rare trace occurrences of rectorite. In the Pays de Bray, there is often traces of pyrophyllite and this also sometimes occurs in the Haute-Marne. Chlorite, also in trace amounts, occurs sporadically in the Berriasian of the Weald.

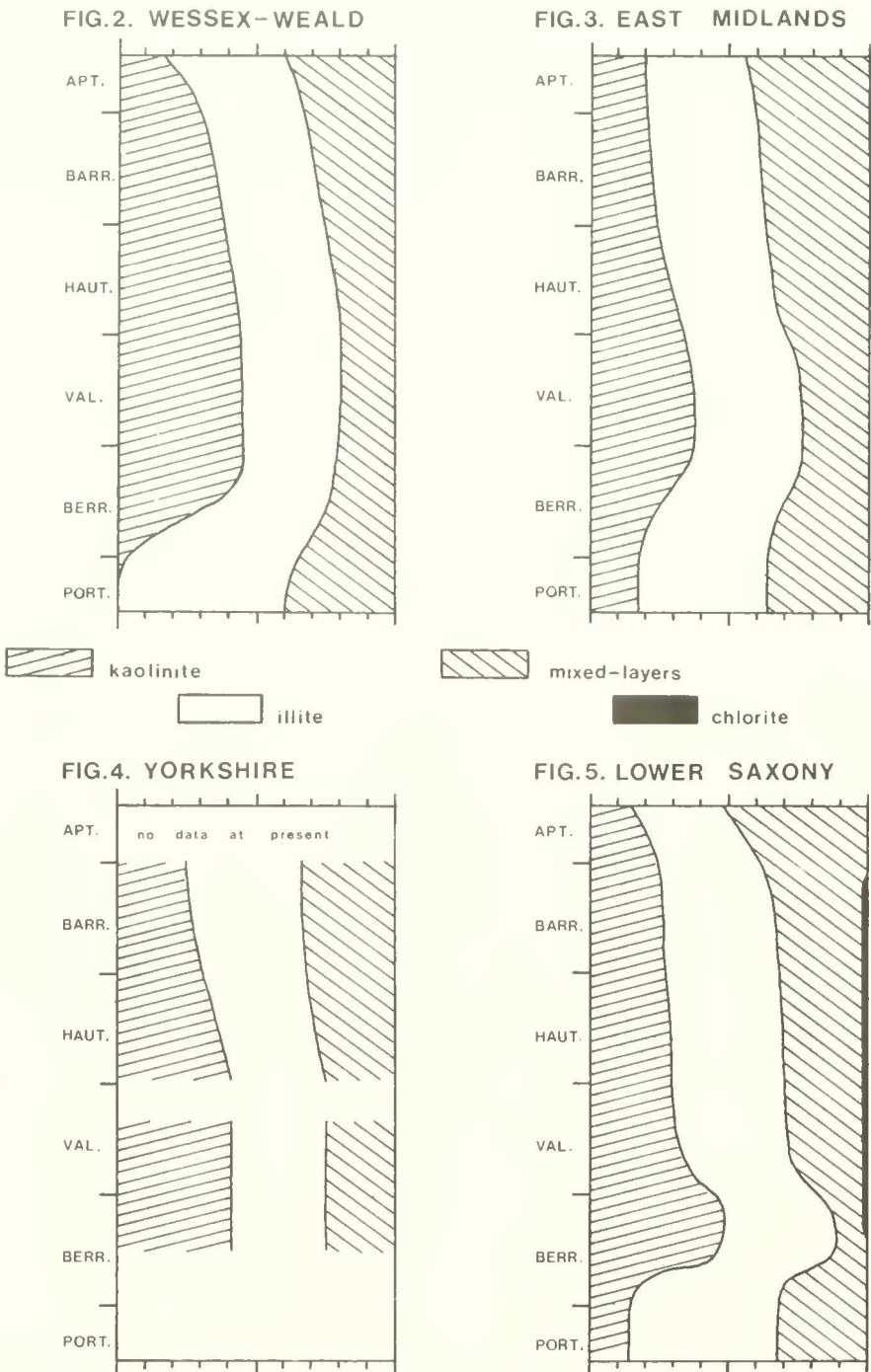
### NORTH OF THE ANGLO-BRABANT MASSIF

Unlike sediments south of the massif, there is no sharp increase in kaolinite and decrease in mixed-layer minerals where the Jurassic-Cretaceous boundary is conformable (e. g. Lower Saxony and Bornholm, Figs. 5 & 9). However, where there is considerable Portlandian and Berriasian missing, there are markedly higher amounts of kaolinite and less mi-

xed-layers in the Cretaceous than in the Jurassic (e. g. the Yorkshire Basin and Moray Firth Basin, Figs. 4 & 6).

The mixed-layer minerals are dominated by fairly randomly interstratified 10–14Å illite-smectites everywhere except Lower Saxony. In Lower Saxony, Al-hydroxy vermiculites, often with illite interlayering, are common from the Berriasian to late Barremian. They were both preceded and suc-

ceeded by illite-smectites. Other mixed-layer minerals are generally absent apart from an isolated, but noteworthy, occurrence of poorly interstratified kaolinite-smectite in the Barrenian Gravenhorst Sst. in Lower Saxony. Chlorite is also only found in Lower Saxony where it occurs in trace quantities.



FIGS.2-5. TRENDS IN RELATIVE ABUNDANCES OF CLAY MINERALS FOR AREAS OF NW EUROPE



FIG.6. MORAY FIRTH

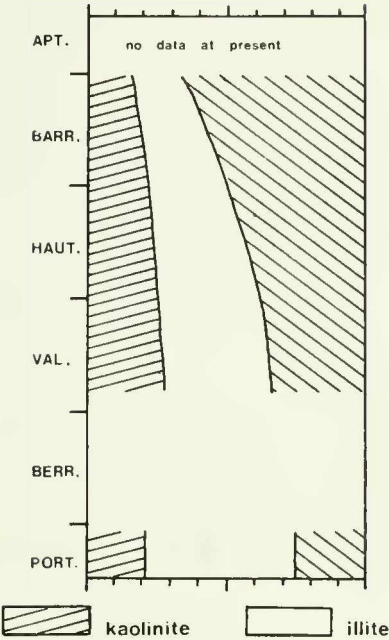


FIG.7. PAYS DE BRAY

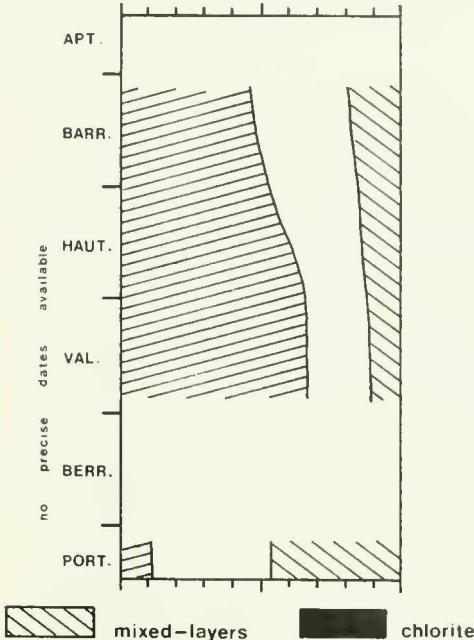


FIG.8. BOULONNAIS

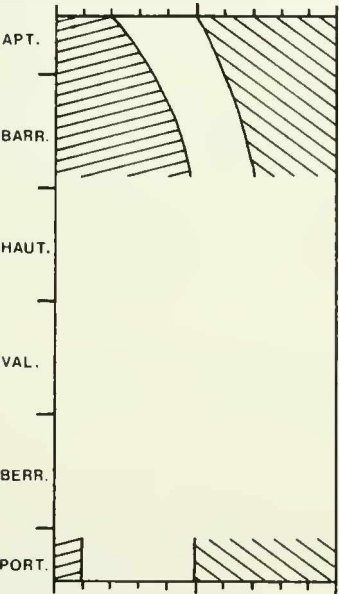
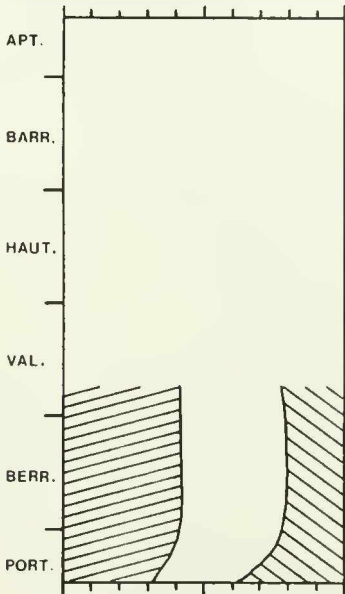


FIG.9. BORNHOLM



FIGS.6-9. TRENDS IN RELATIVE ABUNDANCES OF CLAY MINERALS FOR AREAS OF NW EUROPE

FIG.10. MONS

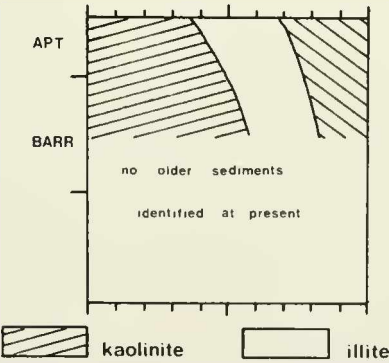
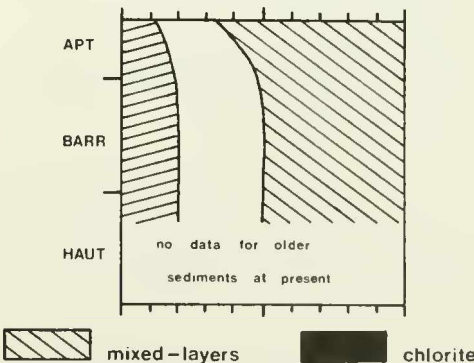


FIG.11. CENTRAL NORTH SEA



FIGS. 10 & 11. TRENDS IN RELATIVE ABUNDANCES OF CLAY MINERALS FOR TWO AREAS OF NW EUROPE

## EARLY CRETACEOUS SOURCE-AREA WEATHERING ENVIRONMENTS DEDUCED FROM CLAY MINERAL ASSEMBLAGES AND TRENDS

### INFLUENCE OF PARENT ROCK COMPOSITION

Reconstruction of the palaeogeology in the Early Cretaceous indicates that the source massifs surrounding the sedimentary basins had cores of Palaeozoic sediments flanked by Mesozoic sediments (ALLEN 1967, 1975, 1981). Igneous and metamorphic rocks were mostly confined to the cores of the massifs. The mineralogy of these rocks has been described by SLADEN (1980 & in press) and ALLEN (1967). They show that the cores were dominated by illite (mostly the 2M polymorph) and chlorite, whilst the flanks comprised mixtures of illite, kaolinite, mixed-layer minerals and chlorite. All clastic sediments were rich in quartz and fairly low in feldspar; carbonate rocks were scarce.

Sedimentological and stratigraphic studies indicate that, in general terms, the massifs were rapidly uplifted during the Berriasian and then gradually submerged, particularly in the Hauterivian, Barremian and Aptian (ALLEN, 1967, 1975, 1981; KEMPER 1973 a, b & c; KAYE, 1964). The submergence of the massifs resulted in overstepping of the earlier Mesozoics, with the Cretaceous slowly coming to lie upon older Palaeozoic rocks. Gradually then, the extent of kaolinite-bearing source rocks would have diminished. This change in source rock mineralogy cannot however produce the trends (and assemblages) of clay minerals because:

1. the content of 2M illite throughout the late Portlandian to early Aptian sediments is always very low or often zero, whereas the source rocks, in particular the Lower Palaeozoic, had considerable 2M illite.
2. chlorite is usually absent or, if present, it only occurs in trace amounts, whereas chlorite is fairly common in nearly all the possible source rocks.
3. kaolinite-rich sediments occur in areas where kaolinite-rich sources would have been either very minor or non-existent e. g. the Mons Basin which was surrounded by mostly Lower Palaeozoics rich in illite and chlorite.
4. the late Portlandian and very early Berriasian contain much less kaolinite than the earlier Mesozoics which, at the time, were the main source rocks according to HOWITT (1964) and ALLEN (1967, 1975).

The lack of correlation between the clay mineralogy of source massifs and that of the sediments indicates that extensive changes in clay mineralogy occurred in the soils and saprolites that covered the massifs. The nature of this mineralogical change between source rocks and their soils and saprolites can be used to provide considerable insight into the weathering environment, as shown below.

### WEATHERING ENVIRONMENTS IN LATE PORTLANDIAN AND EARLY BERRIASIAN SOURCE AREAS

Most of the late Portlandian and early Berriasian contains less kaolinite and chlorite and more illite-smectite and illite

zoics. This difference could be the result of (a) disintegration of muscovite flakes into smaller-sized particles of illite and, (b) neoformation of illite-smectites and illites. Although the breakdown of muscovite must have been occurring, the process could not have been of great importance because it was illite-smectites, not illites, that were major products. The evidence therefore suggests considerable neoformation of illite-smectites and possibly illites, in soils and saprolites in the source-areas.

Today, the neoformation of illite-smectites and illites is commonly recorded in warm, arid and semi-arid areas e. g. S Europe and N & NE Africa (DIXON & WEED, 1977). In these regions, the rainfall is under 1000 mm, soil moisture moves mainly upwards and cations are retained and accumulate creating alkaline profiles ('pedocal'). Carbonates become concentrated close to, and often at the surface, whilst organic matter is oxidised and as a result, organic acid production is low. Similar conditions can be envisaged in the late Portlandian and early Berriasian source-areas and they agree well with other environmental evidence which indicates very low relief around the basins and a warm semi-arid 'subtropical' environment with very high evaporation, at times sufficient for evaporites to form (HOWITT 1964; SCHOTT 1950; WEST 1975). The gradual increase in kaolinite through the late Portlandian and early Berriasian can be interpreted as a reflection of decreasing aridity and increased leaching in source-areas which were gradually being uplifted. These changing conditions slowly introduced a weathering environment in which kaolinite was more likely to be both produced, and preserved.

### WEATHERING ENVIRONMENTS IN SOURCE AREAS BETWEEN THE LATE BERRIASIAN AND EARLY APTIAN

Over large areas of NW Europe, weathering in this period occurred on rocks dominated by illite and containing lesser amounts of kaolinite, chlorite and mixed-layer minerals. The products of the weathering environment were soils and saprolites containing higher amounts of kaolinite, less illite and usually no chlorite. Mixed-layer minerals were usually characterised by the development of either Al-hydroxy vermiculites or fairly randomly interstratified illite-smectites.

The mineralogical changes which took place require acidic and leaching environments. The quantity and type of mixed-layer minerals produced indicates that they were the weathered products of illite, chlorite and other mixed-layer minerals. The weathering of chlorite must have contributed some mixed-layer minerals but most of the Al-hydroxy vermiculite is acid insoluble and therefore formed from illite and 'illitic' mixed-layer minerals as opposed to chlorite (RISTORI et al. 1974). Al-hydroxy vermiculite and illite-smectite develop from illite in soils by leaching of interlayer potassium. The abundance of random interstratifications and similarity in particle size of the mixed-layer minerals and illite indicates that leaching of potassium occurred along certain layers of the illite lattice ('layer weathering').



On many areas of the massifs, the conditions favoured the formation of Al-hydroxy vermiculite e. g. the Anglo-Brabant and Rheinische Massifs which sourced much of the Weald and Lower Saxony. Field studies and laboratory experiments have shown that fairly intensive acid leaching conditions are required to form this mineral with a pH around 5 and frequent wetting and drying (RICH 1958, 1968; DOUGLAS 1965; FEIGENBAUM & SHAINBERG 1975; VICENTE et al. 1977). The present-day soils where these conditions prevail are podzols and Al-hydroxy vermiculite is frequently found in them (KLAGES & WHITE 1957; JACKSON 1959; MILLOT 1970; DIXON & WEED 1977). Similar podzolic profiles can be envisaged covering large areas of the massifs particularly during the late Berriasian and Valanginian when Al-hydroxy vermiculite was most common.

The less common mixed-layer minerals that developed also indicate acidic and leaching conditions. Vermiculite-smectite can be found in present-day podzols (GJEMS 1963). Beidellites and complex chlorite-smectites are found today in poorly-drained acid soils on rocks containing illite and chlorite (DIXON & WEED 1977). In the Early Cretaceous, these would have formed on areas of poorly-drained Lower Palaeozoic e. g. around the Mons Basin. Kaolinite-smectite which is present in Lower Saxony is also found forming today in acid profiles (pH 6.5–5.0) under a warm humid climate (ALTSCHULER et al. 1963) and this suggests that these conditions existed on part of the Rheinische Massif. The few recent reports of pyrophyllite forming in soils are from podzolic profiles on rocks rich in primary aluminium silicates in areas that are humid with hot summers (DIXON & WEED 1977). These conditions may be envisaged for parts of the Armorican Massif and the

Vosges. The pyrophyllite that developed then found its way into the sediments of the nearby Pays de Bray and Haute-Marne.

The development of acid and leached 'podzolic' profiles over large areas of the source massifs seems certain and the climatic implications that this has can be compared to other lines of evidence on the climate. Palaeospastic reconstructions indicate a position about 30–35°N whilst palaeobotanical, geochemical and sedimentological studies indicate a warm humid-temperate climate (equivalent to humid subtropical of some authors). Sound evidence exists to show that conditions were warm, frequently with high humidities, and seasonably wet, although the seasons could have lasted for very different periods than today's (e. g. ALLEN et al. 1973; ALLEN 1975; ALVIN et al. 1981; BATTEN 1975; SAKS 1975, VACHRAMEEV 1978). Podzols can be found forming today in warm humid-temperate climates that exist in mid-latitudes near the edges of continental land masses that are close to large oceans (e. g. BRIDGES 1970). The conditions in these areas can be summarised as follows: hot humid summers and short mild winters, rainfall 1250–1500 mm per annum and fairly evenly distributed, average temperature of coldest month around 5°C and warmest month around 25°C. The existence of similar conditions to these satisfies both the clay mineral features observed here and the climatic evidence of earlier studies (op. cit.). Clearly, NW Europe lay close to large 'oceanic' water masses with Tethys to the south, the opening Atlantic to the west and the Boreal ocean to the north. The high rainfall would have offset the effects of evapotranspiration in the warm temperatures and maintained a leaching environment in the soils and saprolites.

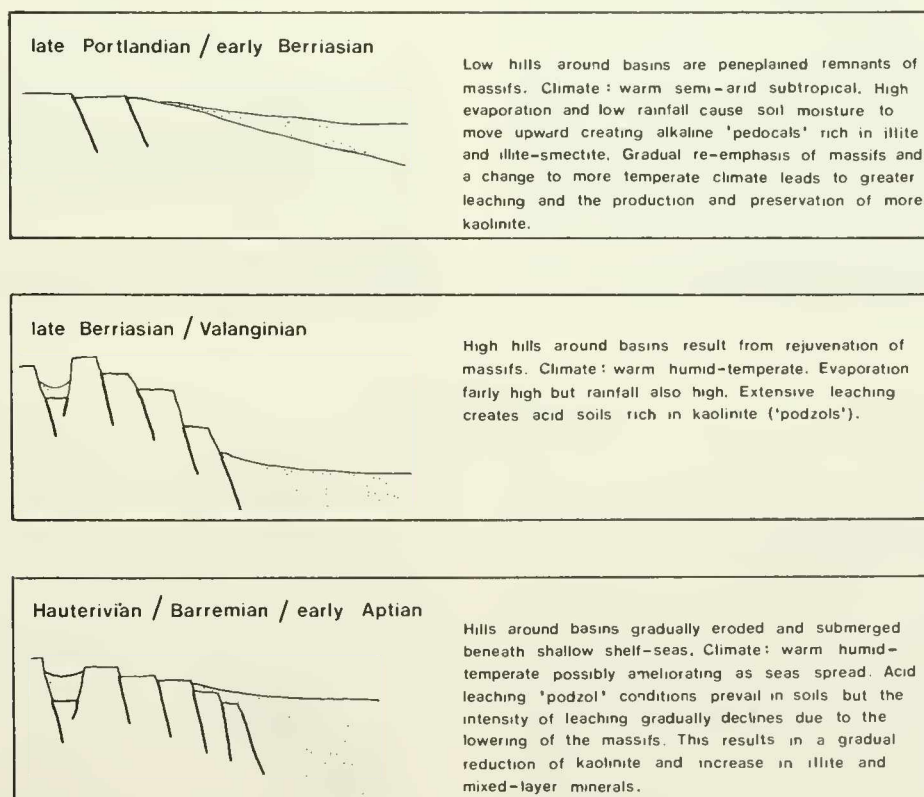


FIG. 12. SUMMARY OF CHANGES IN CONDITIONS ON SOURCE-AREAS DURING THE EARLY CRETACEOUS IN NW EUROPE

(At the same time, the high evapotranspiration would have created the high humidities that have been recognised). The abundance of coniferous foliage that grew, together with siliceous parent rocks, would have helped to establish acidic conditions in the leached profiles. Gibbsite and other aluminium hydroxides which form in intensively leached soils have not been found. Their formation seems unlikely because they favour parent rocks low in silica and tropical climates.

Studies of recent podzolic profiles have shown that Al-hydroxy vermiculite is characteristic of more intensively leached profiles than illite-smectites and smectites and also that kaolinite is more likely to form from mixed-layer minerals as the intensity of leaching increases (BROWN & JACKSON 1958; GJEMS 1963; ROSS & MORTLAND 1966). Consequently, high contents of kaolinite, together with Al-hydroxy vermiculite, indicate more intensively leached podzols than those with illite-smectite and lower contents of kaolinite. This feature is clearly relevant to Early Cretaceous source massifs because, from the Hauterivian to early Aptian kaolinite content declined and Al-hydroxy vermiculites gradually disappeared whilst illite-smectites occurred more frequently. These changes must therefore reflect gradually less intensive leaching on the massifs. Now,

at the same time, the massifs were gradually being submerged and this suggests that their topography and elevation was significant in determining the extent of leaching. It follows that during the late Berriasian and Valanginian, when massifs were relatively high, leaching was fairly effective and so kaolinite and Al-hydroxy vermiculite formed fairly easily. From the Hauterivian to early Aptian, the massifs were being transgressed and lowered. Leaching became less effective and so illite-smectites tended to develop together with smaller amounts of kaolinite (Fig. 12). However, there may also have been some climatic amelioration during this period caused by the spread of the oceans and the general lowering of the massifs.

The correlation between kaolinite content and type of mixed-layer mineral with leaching intensity on the massifs can also explain some of the local changes in clay minerals. For example, in the Weald during the Valanginian, the occurrence of illite-smectite and low kaolinite is seen to correlate with the argillaceous sequences which ALLEN (1975) has showed were deposited when the massifs were relatively low. Conversely, Al-hydroxy vermiculite and high kaolinite correlates with the more arenaceous sequences which developed when the massifs were relatively high and therefore more easily leached.

## CONCLUSIONS

The clay minerals in many Early Cretaceous fine-grained sediments in NW Europe have often remained unaltered since transportation from their source massifs. They closely reflect the composition of soils and saprolites in the source-areas and can be used to reveal features of the source-area weathering environment.

The trends and assemblages of clay minerals reinforce other geological data on the nature of the source-areas. They indicate that from the late Portlandian through the Berriasian and into the Valanginian the block-and-basin topography that existed was gradually re-emphasised and NW Europe 'drifted' from a warm semi-arid subtropical climatic zone into a warm humid-temperate zone. Then, from the Hauterivian to the early Aptian, the relief was gradually lowered and large parts of the massifs were submerged beneath shallow seas.

The climate essentially remained warm humid-temperate but may have suffered some amelioration due to the development of large areas of open sea.

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