

ORIGIN AND CLASSIFICATION OF COASTAL PLAIN KAOLINS, SOUTHEASTERN USA, AND THE ROLE OF GROUNDWATER AND MICROBIAL ACTION

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Abstract—Along the inner Coastal Plain, kaolinite-metahalloysite-rich, neritic muds of Cretaceous-Eocene age have undergone intense postdepositional alteration in the recharge area of the regional groundwater system. Weathering processes have had the following profound effects on the original sediments: 1) strong compositional and textural modification of both clay and non-clay minerals; 2) whitening of the originally darker sediments by partial removal of organic matter, Fe and Mn; and 3) recrystallization of kaolinite and metahalloysite, most conspicuous where there are coarse stacks and vermiforms. Where the combination of initial sediment composition and alteration intensity was most favorable, these changes have produced important deposits of commercial quality, which now sustain the world's largest kaolin production district. The earliest change was partial sequestration of iron as sulfide and concurrent destruction of some organic matter, mediated by sulfate-reducing bacteria. Subsequent weathering resulted in gradual leaching of alkalies, alkaline earths, iron and silica, and attendant nucleation and growth of minerals compatible with the compositional changes. The existence of several closely spaced erosional unconformities, separated by neritic sediments, is proof that weathering conditions commonly changed at a given site, in response to changes in thickness or lithology of the overlying rocks. Dysoxic \rightarrow \leftarrow oxic reversals modified both the rate and kind of alteration. ("Dysoxic" refers to molecular oxygen concentration too low to be toxic to anaerobes or cause abiotic oxidation; less extreme than "anoxic".) Kaolins were produced partly by slower dysoxic weathering in saturated groundwater zones but mainly by more rapid oxic weathering in unsaturated zones, where bauxites also locally formed. Gradual transformation of some sediments to kaolin rarely began and ended in the same epoch. At several places most of the kaolinization (see "Definitions") took place during Recent time, tens of millions of years after deposition of the sediments. Since the kaolins resulted from postdepositional alteration rather than sedimentary processes, they are better referred to as "Coastal Plain" rather than "sedimentary" kaolins.

Key Words—Bauxite Origin, Kaolin Classification, Kaolin Origin, Microbial Weathering, Weathering.

INTRODUCTION

The early interpretation of a sedimentary origin for these kaolins was suggested by their megascopic features and geologic occurrence in sedimentary rocks (Ladd 1898; Veatch 1909; Smith 1929; LeGrand and Furcron 1956; Kesler 1963). It drew support from several assumptions now known to be untenable and did not consider important factors like the regional groundwater system, the genetic relationship between associated kaolins and bauxites and the key role of microbially mediated reactions. These factors could not be recognized until sufficient petrologic and groundwater information had accumulated, development of electron microscopes enabled petrographic study of clays and research in microbiology revealed the high microbial populations and chemical reactions they mediate in Coastal Plain rocks (Chapelle et al. 1988; Chapelle and Lovley 1990; Chapelle 1993; Ehrlich 1996).

Detailed studies of chemistry, mineralogy and petrology began with the work of Jonas (1964), who studied oriented thin sections of kaolins near Dry Branch, Georgia. He reported that large (kaolinized) muscovite platelets tend to conform to gross stratifi-

cation, while kaolinite crystallites do not, a clear inconsistency with sedimentary origin. He concluded that much of the original sediment was not kaolinite and that the present kaolins formed by postdepositional changes.

Detailed mineralogic and petrologic studies of the kaolins began in the 1960s and 1970s, when electron microscopes became generally available (Bohor and Randall 1971; Austin 1972; Patterson and Buie 1974; Tschudy and Patterson 1975; Hurst et al. 1979; Rogers 1979; Barker et al. 1982; Hurst and Rigsby 1984; Barker 1985; Jones 1988; Hurst and Pickering 1989a, 1989b; Pickering and Hurst 1989; Melear 1990; Al-Sanabani 1991; Lowe 1991; Dodge 1991; Barker and Hurst 1992, 1993). Extensive face-to-face association of kaolinite platelets in younger (hard) kaolins, which have undergone the least postdepositional change, indicates a neritic origin which is attested to also by the relative abundance of pyrite and by the extensive neritic trace fossils in these kaolins (Schrod-er 1982; Webb and Sprague 1990).

As investigations progressed, they suggested other basic inconsistencies with the early idea that the kaolins formed as they are today by direct sedimentation.

For example, if these extensive kaolins could form during the 30-my Cretaceous-Eocene interval, why did none form during the succeeding 30-my interval? Clearly, kaolinitic detritus continued to form and erode from the same Piedmont source area, was transported as before and periodically sedimented in similar depositional environments. In other tropical and subtropical areas, as well, kaolinitic detritus was generated and sedimented in the same way as during Cretaceous-Eocene time, but did not produce sedimentary kaolin. No modern sedimentary kaolin is known to be forming today.

During a century of commercial practice and experimentation with kaolin slurries, possibilities for removing deleterious impurities from kaolin by suspension and deposition have been thoroughly explored. Aside from the degritting of coarser particles, the only significant improvements that have been achieved involve synthetic dispersants/flocculants, electromagnetic separation, acid leaching, calcination or similar methods, none of which is part of natural sedimentation. These beneficiation processes have not produced commercial-grade kaolin from neritic clay by simulation of natural sedimentary processes. The cited inconsistencies and the inability to experimentally produce kaolin by sedimentation alone strongly suggest that more than sedimentation was involved and that origin by direct sedimentation is not possible.

Sedimentary processes are not thought capable of producing a commercial kaolin for 2 well-known reasons: (1) Weathering, which produces kaolinite particles simultaneously, produces hydraulically equivalent particles of smectite, illite, illite/smectite and ferric pigments. During their transport and deposition, organic matter and biogenic silica generally are added. These intimately associated components cannot be separated efficiently by sedimentary processes. (2) Surface forces strongly influence the behavior of suspended clay-size particles and inhibit their hydraulic separation. These 2 facts account for the inability of sedimentary processes to make a commercial kaolin.

Garrels and Mackenzie (1971) and others have observed that nearshore marine muds derived from similar source areas are about the same everywhere, chemically and mineralogically. A century of extensive geologic investigation has shown that marine muds like those in the coastal plain of the southeastern United States do not contain kaolin anywhere except where they have been subjected to strong postdepositional alteration.

Only 2 natural processes are known to be capable of altering clays or any other aluminosilicate rock into kaolin: weathering and hydrothermal alteration. Isotopic measurements verify that these clays crystallized at low temperatures (Hassanipak and Eslinger 1985) and thus are products of weathering. If they are sedimentary in origin, their composition must be essen-

tially the same as it was in the initial sediments. The point to be clarified is whether the minerals, textures and bulk compositions of these kaolins are attributable to suspension and deposition or to postdepositional weathering.

DEFINITIONS

Wide usage by many individuals with diverse backgrounds tends to blur the meaning of often-used words. To ensure the clear meaning of key words used in this paper, a few definitions are offered.

Kaolin

The shortest definition consistent with general usage is a light-colored, clayey rock consisting predominantly of one or more kaolin minerals. Most kaolins are predominantly kaolinite, often with metahalloysite, but also may be predominantly halloysite or dickite. Mappable nacrite deposits have not been found. The color of kaolin ranges in value from white to almost black and in hue from pink to yellow-brown or red-brown. It can be soft, earthy or hard—even brittle and nonplastic.

The percent of kaolin minerals in crude kaolins ranges widely, from about 85–95% in the Coastal Plain kaolins of the southeastern United States and the Rio Jari kaolins of Brazil to less than 20% at Cornwall, England, and Hirschau, Germany. Not only crude kaolins but also many different products manufactured from them are all called “kaolin”. While “kaolin” is a rock name, it originated as and still is mainly a trade name. Specimens in museums and repositories are atypical of most crude kaolins.

Kaolinization

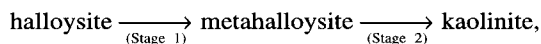
“Kaolinization” is the formation of kaolin by the weathering of aluminosilicate minerals (Gary et al. 1974) or clay minerals (resilication of gibbsite to kaolinite, recrystallization of metahalloysite).

Kaolinite-metahalloysite

Transmission electron microscopy (TEM) observation of hundreds of samples from kaolins in Cretaceous-Eocene rocks in Georgia and South Carolina has revealed that metahalloysite is a conspicuous phase, except where kaolinization has been sufficient to alter it to kaolinite. Though its presence in these kaolins has been reported several times (Austin 1972; Hurst et al. 1979; Rogers 1979; Pickering and Hurst 1989; Hurst and Pickering 1989a, 1989b; Lowe 1991), its prevalence has not been recognized heretofore. X-ray diffraction (XRD) does not reliably distinguish it from kaolinite. For simplicity in this paper, the essential kaolin mineral is called “kaolinite” when metahalloysite is scarce or not seen with TEM, and is called “kaolinite-metahalloysite” when both phases are clearly present.

Metahalloysite

This is the intermediate phase in the paragenetic mineral sequence:



observable with TEM. During the first stage, irreversible dehydration does not change the halloysite habit but shifts components of the structure and thus produces another phase, metahalloysite. During the second stage, recrystallization of metahalloysite destroys its habit while kaolinite crystallites nucleate and grow. The end product is a striking "saw-toothed" elongate of kaolinite platelets (Lowe 1991; Hurst 1997).

Clay

The definition recently adopted by The Clay Minerals Society (CMS) and Association Internationale Pour L'étude des Argiles (AIPEA) (Martin 1991; Guggenheim and Martin 1995) differs from those long-recognized by geological societies, engineering societies, the U.S. Department of Agriculture and the International Society of Soil Science, which are much larger groups (for a comparison of definitions, see Gary et al. 1974). Many in these groups are also CMS members but might be unaware of the newly adopted definition. As used in this paper, "clay" means the common denominator of the definitions of all these groups: a pulverulent material consisting of particles and/or crystallites finer than 74 μm , the approximate limit of visual resolution, and essentially without compositional connotation. For engineers, the maximum particle/crystallite size is 74 μm ; for geologists, the maximum is 4 or 2 μm , depending upon grade scale (Pettijohn 1949); for soil scientists, 2 μm ; for CMS and AIPEA, about the same but unspecified, and with a qualified compositional connotation.

GEOLOGIC SETTING

Inner Coastal Plain sediments range in age from Late Cretaceous to Holocene. The wedge of sediments is eroded out at its NW edge, the Fall Line, and thickens seaward at the rate of 5 to 10 m km^{-1} (Figure 1). Its lower surface is a pronounced unconformity, concave upward and steepening westward, over metamorphic and igneous rocks. Its upper surface, the present weathering surface, is youthful where dissected by streams but old in interfluvial areas exposed to uninterrupted weathering since a recession of the sea 15 my ago. The upper 3 major erosional surfaces have similar relief and drainage patterns (compare Figure 14 in Hurst et al. 1966 with a modern topographic map). The fourth erosional surface, on the metamorphic and igneous basement, is steeper than the others near the Fall Line.

During the Cretaceous-Eocene interval, world sea levels were consistently higher than now, at times by

several hundred ft. Climate was warmer or about the same as today (Hassanipak and Eslinger 1985). Each major transgression left a retrogradational parasequence set, a fining-upward sequence. Kaolin bodies have been found, locally, in the upper parts of at least 3 such sets. Each time the sea receded, uppermost rocks were exposed to oxic weathering and erosion. Several retrogradational sets were deposited, although generally no more than 2 or 3 are exposed at a given outcrop.

Gross lateral and vertical variations in lithology and permeability of the host-rocks relate partly to facies changes and unconformities. Likewise, some gross variations within the kaolins relate to heterogeneities in the host sediments, but most variations relate to the kind, intensity and duration of postdepositional processes described below.

EARLY CONSIDERATIONS OF ORIGIN

The long-held idea that kaolins in the Coastal Plain are sedimentary kaolins has depended from the start upon a tacit assumption that sedimentary processes can make a kaolin. This idea gained wide acceptance while the focus of inquiry was on visual observation, but it has drawn support only from assumptions and observational inferences. For example, from the presence of kaolins in a sedimentary sequence and relict bedding in some kaolins, a sedimentary origin was inferred. Kaolinite in the kaolins was assumed to be the same as that deposited. The presence of apparent coarse clasts of kaolin in some units was regarded as strong evidence that kaolins existed prior to deposition of the clasts. Actually, some of these coarse angular masses are kaolin clasts, while others are clay clasts altered subsequently to kaolin. Accumulated evidence now shows that these assumptions and inferences are untenable.

SYNOPSIS OF NEW EVIDENCE BEARING ON ORIGIN

A plot of the distribution of kaolins in relation to the southeast-dipping Coastal Plain strata shows that kaolins are restricted to the fresh water recharge area of the regional groundwater system (Figure 1).

TEM-assisted petrological studies by the authors and associated students during the last decade show that nearly all inner Coastal Plain rocks (sands and clays alike) bear evidence of strong postdepositional alteration, except those that are least permeable and were never exposed to oxic weathering.

Extensive mining and drilling have disclosed that most kaolins are in the upper part of fining-upward sequences deposited during sea highstands. Significantly, fewer than 5% of the clays in this stratigraphic position are now sufficiently pure, white and continuous enough to be commercial kaolin—only those that

Index map showing location of cross-section A-A', in Central Georgia.

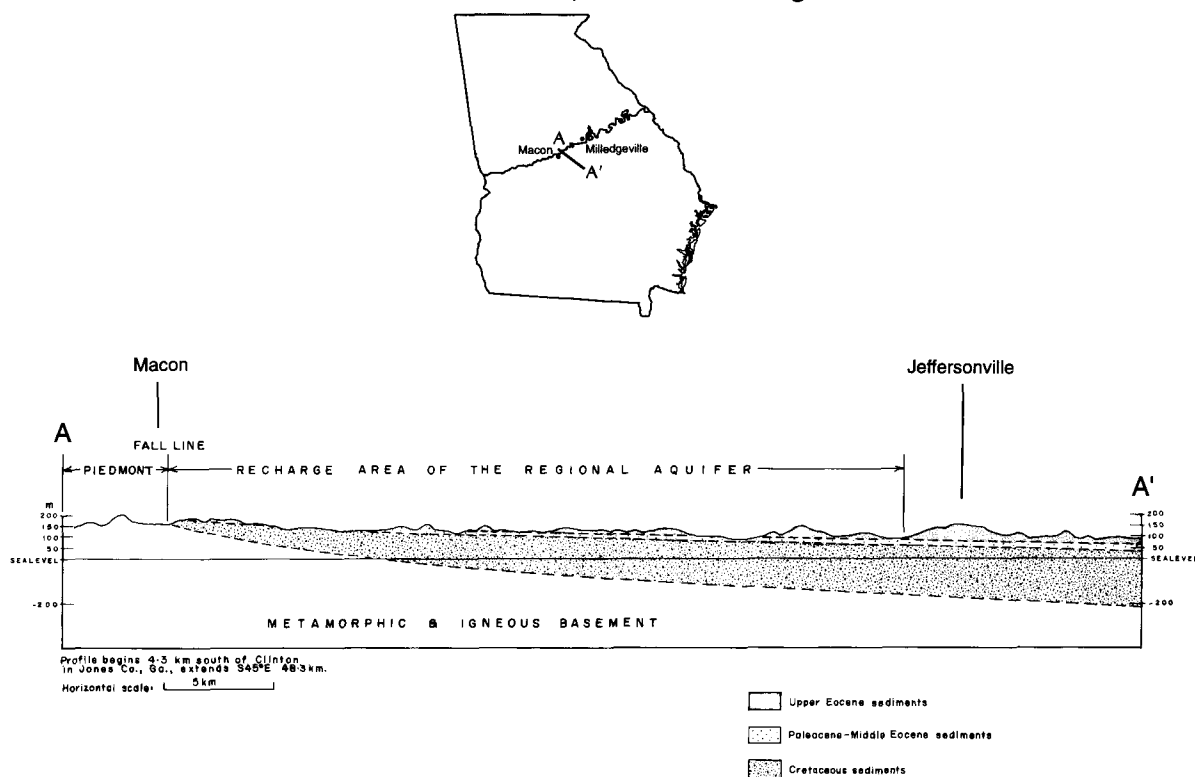


Figure 1. NW-SE profile of the recharge area of the Coastal Plain aquifer in central Georgia. All represented surfaces are erosional unconformities. The uppermost surface is accurate. Three underlying surfaces (dashed) are approximations. The base of Upper Eocene and the top of Cretaceous sediments are smoothed curves through 10 data points, approximations from Hetrick and Friddell (1990). The surface over metamorphic and igneous basement is an approximation based mainly upon information in Cramer (1974). Actual relief of the dashed surfaces is probably 15–40 m, as measured by Hurst et al. (1966) in one updip area. This estimate of relief is consistent with measured elevations of kaolins in Tertiary sediments, ≈ 148 –62 m, and in Cretaceous sediments, ≈ 75 –50 m.

bear evidence of having undergone intensive alteration.

Field and laboratory studies have shown that compositional variations in the kaolins relate not so much to differences in source area and depositional environment as to location within the groundwater system and differing degrees of kaolinization and diagenesis, which most of the time relate to age. Variations in the lithology of the source area are detectable in studies of heavy minerals (Hurst 1952–54, unpublished) and trace elements (Dombrowski and Murray 1984; Dombrowski 1992, 1993),

Information about the abundance of subsurface microorganisms in this region and the importance of microbial mediation in weathering reactions that produced the kaolins has become available mostly during the last decade (Chapelle et al. 1988; Chapelle and Lovley 1990; Chapelle 1993; Ehrlich 1996).

The same postdepositional processes that produced kaolin also produced bauxite where alteration was suf-

ficiently intense and prolonged. In middle Georgia and South Carolina, the bauxites are thin or spottily distributed, but farther west, as near Andersonville, Georgia, they are extensive. Hydrolytic weathering of aluminous rocks produces kaolin minerals first, then increasing gibbsite as early kaolin minerals are desilicated. This kinship of kaolins and bauxites was not recognized by early proponents of “sedimentary kaolin”.

SYNOPSIS OF EVIDENCE FOR ORIGIN BY POSTDEPOSITIONAL ALTERATION

These kaolins (and associated bauxites) are located on what has been a passive continental margin for the last 100 my. A humid subtropical climate has prevailed during most of that time, as shown by paleoweathering profiles. Although paleo-lateritic weathering profiles were formerly thought to have occurred in the southeastern United States, exhaustive search has revealed that none are in the inner Coastal Plain, al-

though Podzolic paleo-profiles indicative of humid subtropical conditions are common (Hurst 1980). The profiles characteristic of humid subtropical and humid tropical weathering are described in pages 59–74 (1980). The inevitable consequence of such a setting would be widespread, long-continued and locally intense hydrolytic weathering. This expectation, based upon the geology and hydrology of the region, is strongly supported by the petrology of the kaolins, as revealed by optical and electron microscopic studies (Jonas 1964; Hurst et al. 1979; Rogers 1979; Grim 1982–85, unpublished; Hurst and Pickering 1989a, 1989b; Pickering and Hurst 1989). How the original sediments have changed in chemical composition and mineralogy is evident from relicts of some sedimented phases and from secondary minerals formed *in situ*.

Twelve important postdepositional alteration sequences are clearly observable or can be reliably deduced. Most of these are evident from electron micrographs in previous publications (Austin 1972; Hurst et al. 1979; Rogers 1979; Jones 1988; Barker 1988; Hurst and Pickering 1989a, 1989b; Pickering and Hurst 1989; Lowe 1991).

1) The earliest postdepositional change at most places was bacterially mediated stripping of Fe from organic matter, kaolinite-metahalloysite, illite and other minerals by HS^- , following sulfate reduction by bacteria, and sequestration of much of the Fe as sulfide. How pyrite (or its precursor) forms in sediments is well known from the work of Jorgensen (1982), Berner and Raiswell (1983), Westrich (1983), Berner (1984), Chapelle (1993), Ehrlich (1996) and others. The amount of pyrite can be limited by rates of supply of decomposable organic matter, reactive Fe and dissolved sulfate. Sulfate was high in the seawater in which most of the sediments were deposited. Pyrite generally is seen in the kaolins except where oxic weathering is evident, where its former presence may still be apparent from residual limonite nodules.

2) Destruction of organic matter. The presence of bacterially mediated pyrite is evidence that an amount of organic matter was destroyed—usually several times greater than the amount of pyrite that formed (Berner 1984). Additional organic matter would have been destroyed in sediments undergoing oxic weathering. Consistent with this, organic matter is conspicuously greater in sediments subjected to dysoxic groundwater only and least in coarse kaolins produced partly or entirely by oxic weathering.

3) Recrystallization of kaolinite-metahalloysite and coarsening of kaolinite. Coarse kaolins have almost completely recrystallized, while very fine kaolins generally have not. Recrystallization and coarsening are most conspicuous in the older, softer kaolins produced by oxic weathering (Pickering and Hurst 1989; Hurst and Pickering 1989b).

4) Extensive *in situ* weathering of feldspar to kaolinite (Jonas 1964; Grim 1982–1985, unpublished; Pickering and Hurst 1989).

5) Alteration of Ti-bearing minerals to anatase, particularly evident in the hard kaolins.

6) Kaolinization of illite (Austin 1972; Pickering and Hurst 1989). This is a slow transformation, most conspicuous in Cretaceous rocks subjected to intense oxic weathering.

7) Diagenetic transformation of biogenic silica to opal-CT and finally to quartz. Ocean waters are now unsaturated with respect to amorphous silica at all depths and probably have been since Cambrian time, when silica-secreting marine organisms first became common. Thus, inorganic precipitation of silica from seawater has been unlikely since Cambrian time, and even less likely since Late Mesozoic, when diatoms became abundant. The silica in Cretaceous-Eocene sediments originated as detrital quartz and as biogenic precipitates. The latter are readily distinguishable in young sediments at high magnification. With the passage of time or an increase in temperature, biogenic silica tends to migrate (due to its high solubility) and gradually transform to opal-CT (Elzea et al. 1994) lepispheres (Hurst and Pickering 1989a, 1989b). Any opal-CT not removed by leaching gradually transforms to fine-grained quartz. Mitzutani (1970) concluded from experimental work and observation that transformation may be complete in 10^9 years at ambient temperatures. In the inner Coastal Plain, lepispheric silica is abundant in Eocene clays, much less common in Paleocene clays and scarce or not present in Cretaceous clays, consistent with Mitzutani's conclusion.

8) Other important changes are the gradual removal through leaching of alkalis, alkaline earths, silica, Fe and Mn. Alkali was released when feldspar altered to kaolinite, when the layer charge of micas decreased during weathering and when illite and coarser mica were kaolinized. Alkali was removed, because relic grains of strongly altered feldspar but no new alkali-bearing phase are found. Loss of silica is indicated by etching of quartz (Austin 1972), dissolution of biogenic silica, gradual destruction of montmorillonite, common presence of gibbsite in kaolins and local bauxitization of kaolin. The loss of Fe and Mn generally was pervasive and hardly visible but locally was conspicuous where Fe was leached from euxenic zones and where Mn, after transport in solution, was deposited as fine, black lithiophorite.

9) Gradual loss of montmorillonite by groundwater leaching. This mineral is much more susceptible to destruction by leaching than kaolinite (Siffert 1962; Millot 1970). Its breakdown can be very slow in low temperature, saturated groundwater zones or rapid in oxic, well-leached zones. Accordingly, clays in saturated zones generally contain more montmorillonite. All Coastal Plain kaolins still contain a little, usually

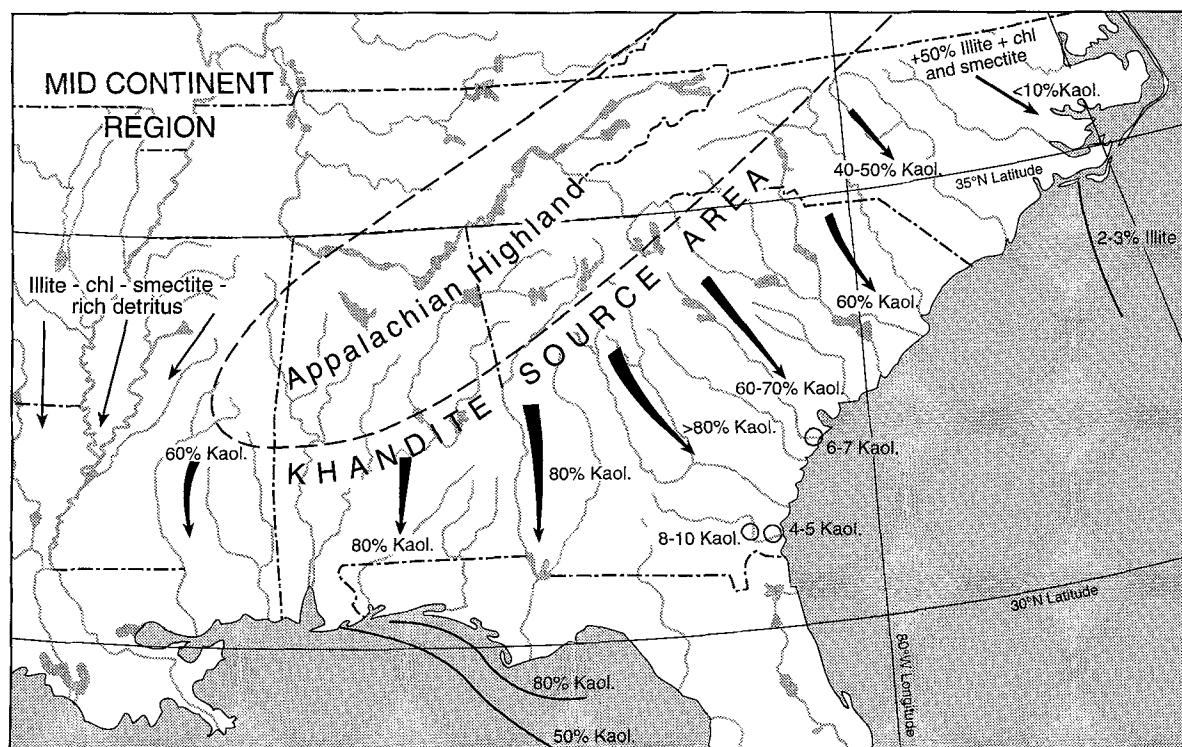


Figure 2. Principal geomorphic features of the kaolinite-rich source area in the southeastern United States, where a humid subtropical climate has long prevailed.

less than 5% but sometimes higher. Clays associated with kaolins but insufficiently altered to have become kaolin commonly contain more than 8% montmorillonite (Millman and Iannicelli 1966). Some commercial kaolins contain up to 50% montmorillonite (Grim 1982–1985, unpublished). Kaolins containing approximately 20% montmorillonite are mined by the Dry Branch Kaolin Co. in Twiggs County, Georgia, as at the Willingham–Birdsey Mine 8 miles south of Macon, Georgia.

10) Slow bacterially mediated deferration by leaching in regional dysoxic saturated zones or in local euxenic zones beneath swamps. Good examples of kaolinization in a perched euxenic zone have been mined in the Sandersville, Georgia, area.

11) Diagenetic changes under long-maintained dysoxic conditions. Pyrite tends to coarsen, making it easier to remove during kaolin processing. Kaolinite tends to coarsen by Ostwald ripening and by open-system recrystallization, but Ostwald ripening is notably slower than open-system recrystallization in well-leached, oxic zones (Hurst 1980).

12) Where rocks become part of the unsaturated zone, as by erosion of cover rocks, weathering proceeds under oxic conditions. The resulting bacterially mediated breakdown of pyrite produces sulfuric acid (Evangelou 1995), which greatly accelerates weather-

ing. Much of the Fe initially present may be removed as sulfate or remain as residual limonite, depending upon local conditions (Blanchard 1968). Oxidation of octahedral iron in kaolinite-metahalloysite assists recrystallization to a purer, better-ordered kaolinite. Oxic conditions favor loss of tetrahedral Al from illite, lowering its layer charge and interlayer potassium. Where a subaerial surface was maintained long enough, a zoned weathering profile developed. These profiles have been found more often in the N-NW portion of the groundwater system rather than seaward, where thicker cover rocks prevented surficial weathering.

SOURCE AREA, COMPOSITION OF INITIAL KAOLINITIC-METAHALLOYSITIC MUDS

Principal geomorphic features in the southeastern United States are shown in Figure 2. Essentially the same patterns of relief and drainage have existed since Cretaceous time. From the northeastern United States down to North Carolina, fluvial drainage now carries illite-, chlorite-, smectite- and quartz-rich detritus to the sea. Similar detritus is transported from the Midcontinent region by the Mississippi River and its tributaries. In contrast, detritus moving seaward from the Appalachian Highland in South Carolina, Georgia, Alabama and eastern Mississippi is dominantly kaolinite-metahalloysite, up to 60% in South Carolina and east-

Table 1. Comparison of generalized compositions of (1) kaolinite-metahalloysite-rich sediment from the Appalachianicola River and (2) Georgia kaolin, wt%, showing differences in mineralogy and chemistry produced by kaolinization.

	Mineralogical			Chemical	
	(1)	(2)		(1)	(2)
Kaolinite-metahalloysite	80–60	95–70	SiO ₂	60–55	53–42
Mica/illite	20–2	10–1	Al ₂ O ₃	30–20	40–36
Smectite	10–5	5–1	TiO ₂	1–2	7–1.5
Quartz	10–1	5–<1	Fe ₂ O ₃	8–4	2–0.3
Biogenic silica	up to 2	<1	CaO	6–2	0.6–0.01
Organic matter	up to a few	<1	MgO	3–1	0.1–0.01
			K ₂ O	3–2	1.2–0.02
			Na ₂ O	1–0.5	0.4–0.05
Accessory minerals	hematite goethite ilmenite rutile others	hematite goethite anatase others	organic carbon	3–0.2	0.1–0.02

ern Mississippi and up to 80% in Georgia and Alabama (Griffin 1962; Blatt et al. 1980; Weaver 1989; Dodge 1991). Climatic patterns of temperature and rainfall induced more intense hydrolytic weathering in this area and thus produced the kaolinite-metahalloysite-rich detritus. It is not diluted by harder-to-weather detritus from the continental interior because of the blocking Appalachian highland. Kaolinite-metahalloysite has been the dominant component of detritus from this source area since Cretaceous time. When it reaches the sea, it flocculates, as in earlier times, and usually settles out within a hundred km of the shore, though sometimes farther. South of the Appalachianicola River, for example, it is the dominant component of bottom muds for 160 km offshore (Griffin 1962).

Considering 1) the composition of less-altered clays, 2) the composition of detritus now moving to neritic environments seaward of the kaolins and 3) the fact that the climate and source area are still about the same as during Upper Cretaceous-Eocene time, the mineralogical composition of muds altered to kaolins in this region can be approximated as kaolinite-metahalloysite 60–80%, mica and illite 5–20%, smectite 5–10%, quartz 1–10%, variable but generally minor feldspar, biogenic silica up to 2%, organic matter up to several percent (geometric average for all muds is 1.1%), accessory ilmenite, hematite, goethite, and other heavy minerals and locally up to several percent of carbonate minerals. Most of the biogenic silica and much of the organic matter in the initial muds probably were added in the depositional environment.

COMPOSITIONAL CHANGES DURING KAOLINIZATION

For a kaolinite-metahalloysite-rich mud to become kaolin, it has to change both chemically and mineralogically. Changes in bulk chemistry take place mostly by the mobilization and translocation of elements, but also by the translocation of fine solids. Mineralogical

changes are possible by degradative changes in a solid, recrystallizational transformations sometimes called “diagenesis” and by mobilization and translocation of a colloidal phase. Changes in mineralogy tend to reflect the composition of the pore fluid because they are particularly responsive to the relative purity and rate of flow of incoming water and to microbial activity (Chapelle et al. 1988; Chapelle and Lovley 1990; Chapelle 1993; Ehrlich 1996). Viable microbes are virtually ubiquitous in sediments. They thrive where suitable energy sources, nutrients and terminal electron acceptors are available, and commonly influence the composition of the pore fluid more than any other factor. By changing the oxidation state of multivalent elements (Fe, Mn and V) and changing the pH of the pore fluid, they enable important changes in bulk chemistry.

The general composition of kaolin and its deduced precursor are compared in Table 1. As the precursor sediment was kaolinized, part of its smectite, illite and silicate was removed. Organic matter, Fe, alkaline earths, alkalis and silica were decreased, while less mobile aluminum was proportionately increased. Considerable organic matter was destroyed by bacterially mediated reactions in the saturated zones and by both microbially mediated and abiotic oxidation in oxic zones, but there was some replenishment by downward movement of dissolved or suspended organic matter. Silica was significantly decreased by leaching. Ferric Fe is insoluble and essentially immobile, except at low pH or in soluble complexes. Whether any of it was removed from oxic zones depended strongly on microbial mediation. Other, usually lesser, multivalent metals whose oxides are insoluble, such as Mn and V, behaved similarly.

DISTRIBUTION OF KAOLINS AND BAUXITES

The outcrop pattern of updip, mainly neritic host sediments and the distribution of kaolin and bauxite

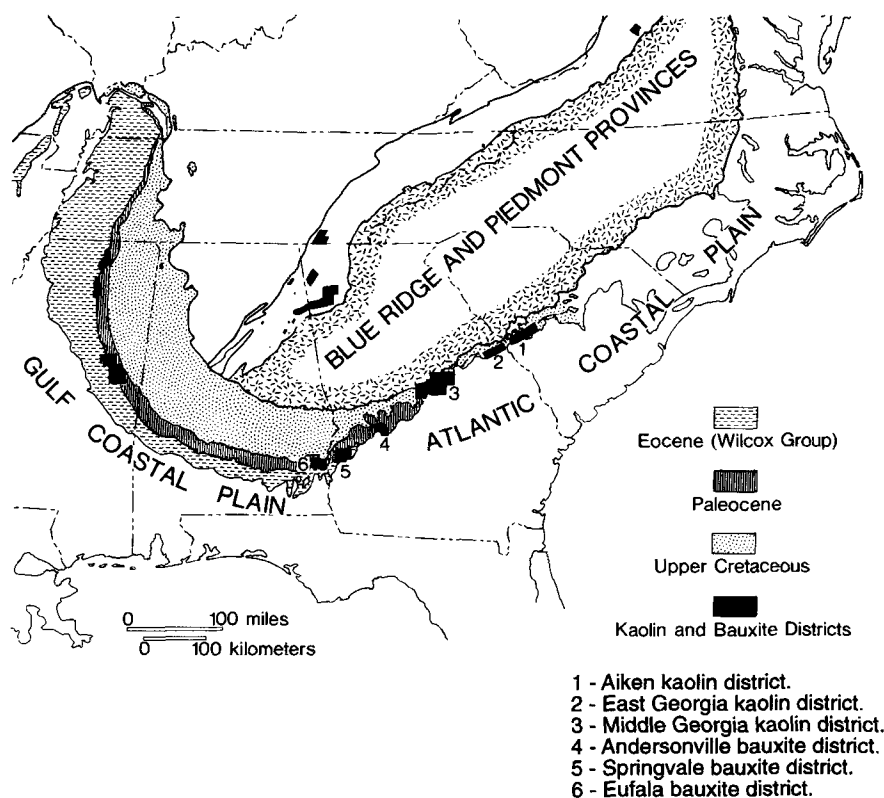


Figure 3. Distribution of Coastal Plain kaolins and bauxites in the southeastern United States.

deposits, are shown in Figure 3. When their distribution is overlaid on Figure 2, they are seen to be restricted along strike to areas where kaolinite-metahalloysite-rich Cretaceous-Eocene detritus was not diluted by harder-to-weather detritus from the continental interior. Transverse to strike, they are restricted to the recharge area of the regional groundwater system, where postdepositional weathering was most intense.

SYNOPSIS OF PHYSICAL-CHEMICAL-MICROBIAL CONDITIONS IN THE REGIONAL GROUNDWATER SYSTEM

The 4 types of environments in a Regional System are: 1) local unsaturated environments, including most soil zones, where lateral water flow is limited; 2) local saturated environments; 3) intermediate saturated environments; and 4) regional saturated environments (Toth 1963; Chapelle 1993).

The overall pattern of groundwater movement in this system (Figure 1) might appear simple, but more complexity is apparent when it is viewed at a smaller scale. Within about 61 m of the surface, groundwater movement is complicated by the surface drainage pattern and strong permeability differences within the strata. Deeper flow is further complicated by 2 major unconformities that transect sedimentary sequences and interrupt lateral continuity. In more permeable lay-

ers, groundwater movement is mostly lateral and sometimes very slow but still not nearly as slow as in confining zones, where water movement is largely vertical. Because of rock heterogeneities, zone interfaces can be quite irregular and an occasional, smaller saturated zone is perched within a larger unsaturated zone.

Unsaturated Zones

These are inhabited by bacteria, viruses, actinomycetes, cyanobacteria, fungi and algae, as well as by microscopic arthropods (Chapelle 1993), but aerobic heterotrophic bacteria dominate, 10^6 – 10^8 cells/g of rock. Generally, oxic conditions prevail. The rates of decomposition of older minerals and crystallization and growth of new phases are greatly accelerated. Leaching gradually removes soluble species while elements like Fe and Mn, whose higher oxidation states are insoluble, are immobilized.

Saturated Zones

Dysoxic conditions prevail and the movement of groundwater can be very slow. Diverse populations of anaerobic and facultative anaerobic bacteria dominate, 10^6 – 10^8 cells/g of rock (Chapelle 1993; Ehrlich 1981, 1996). Microbial processes can deplete organic matter and mobilize Fe by reducing it. Though abiotic reduc-

Bacterial destruction of organic matter and solubilization of Fe in Dysoxic
sediments in Coastal Plain aquifer (after Chapelle & Lovley 1992).

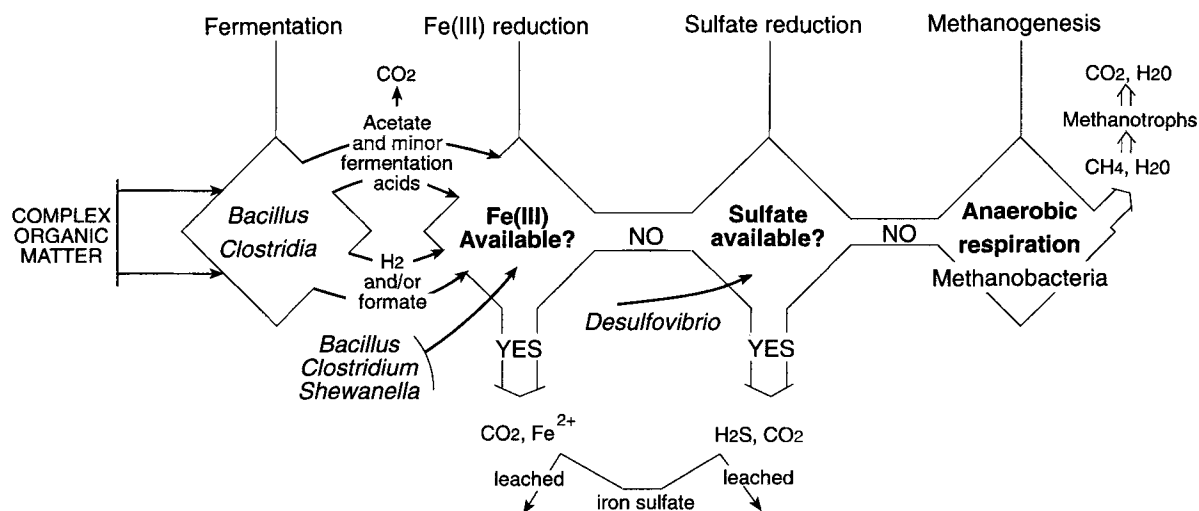


Figure 4. Schematic depiction of carbon and electron flow in dysoxic inner Coastal Plain sediments in relation to destruction of organic matter and mobilization of Fe and Mn during kaolinization.

tion of Fe(III) in hematite, goethite and other ferric minerals is possible, it is inherently much slower than enzymatic reduction. Lovley et al. (1990) have shown that acetate-oxidizing, Fe(III)-reducing bacteria can enzymatically couple the oxidation of organic matter to the reduction of Fe(III) under the conditions typical of deep aquifers. In inner Coastal Plain sediments of South Carolina, they recovered this type of bacteria from several aquifers 20 to 250 m deep where, as expected, they measured high concentrations of dissolved ferrous Fe ($>300 \mu\text{g/L}$). Subsequently, Chapelle and Lovley (1992) found that methanogenesis and sulfate reduction are the predominant terminal electron-accepting processes in sediments containing low concentrations of dissolved Fe. These processes are not important until microbially reducible Fe(III) has been depleted (Lovley and Phillips 1987). Figure 4 is a schematic representation of these relationships. In the absence of reducible Fe(III), respiratory Fe(III)-reducing microorganisms can hardly survive in nitrate-depleted anaerobic zones because they are unable to obtain energy for growth from sulfate reduction, methane production or fermentation. Fermentative microorganisms can metabolize even the organic matter in old sediments, after which Fe(III)-reducing bacteria can couple the oxidation of fermentation products like acetate to the reduction of Fe(III) (Chapelle et al. 1988; Chapelle and Lovley 1990).

The net results of fermentative, Fe(III)-reducing, sulfate-reducing and methanogenic microorganisms are destruction of organic matter; generation of CO_2 ,

H_2S , CH_4 and H_2O ; and solubilization of Fe and other multivalent elements (Figure 4). Where groundwater movement is sufficient, leaching tends to remove these ions as they are released by microbial activity (Chapelle and Lovley 1992).

In subsurface environments, the most fundamental distinction is that between saturated and unsaturated zones. Weathering is very slow in saturated zones and whether organic matter, Fe, Mn and V are depleted depends strongly upon microbial mediation. In unsaturated, oxic environments, on the other hand, weathering is relatively rapid. Organic matter can be destroyed abiotically or biotically, but Fe and Mn become relatively immobile unless complexed. Where rainfall is abundant, the leaching of ions and orthosilicic acid can be fairly rapid.

The 2 types of zones are readily distinguishable by color. Ferrous minerals in saturated zones tend to be pale green when thin enough to transmit light, while ferric minerals like goethite and hematite, which form only in unsaturated oxic zones, are conspicuous by their yellow, brown or red-brown hues. A transition between the 2 zones commonly is indicated by lavender to purple hues, indicating the presence of unhydrolyzed $\text{Fe}(\text{H}_2\text{O})_5^{3+}$ ions. The percent of ferric Fe in oxic zones can be visually quantified with a Munsell Soil Color Chart (Hurst 1977).

IMPORTANCE OF MICROORGANISMS

The importance of microorganisms, particularly bacteria, would be hard to overemphasize. Bacteria are

the most numerous and versatile organisms on earth (Margulis and Sagan 1986; Gould 1996; Brown and Sherriff 1996). They inhabit virtually all surface and subsurface environments (Chapelle 1993). Their importance stems from their ability to enzymatically bypass the high activation energies that retard or block many low-temperature reactions. Also, they drive reactions that are not thermodynamically spontaneous by coupling them in a chain of reactions by which free energy from one or more exothermic reactions in the chain drives a coupled endothermic reaction. Their abundance, ubiquity and proclivity for driving low-temperature reactions that otherwise would not proceed account for their great importance in weathering, without which the kaolins would not have formed. Abundant trace fossils in the kaolins verify that bacteria were involved as early as the period of sediment deposition (Barker 1985; Barker and Hurst 1992). They and other microorganisms are still involved, even in modern processing of kaolin (Eagon et al. 1987). High-brightness kaolin products to be delivered in slurry form are routinely treated with a biocide to control microbe reproduction and meet the industry tolerance level of about 3×10^3 – 10^4 colony-forming units per mL of slurry.

SUMMARY AND CONCLUSIONS

The formation of kaolin requires a higher degree of chemical differentiation than is possible by sedimentary processes. While hydraulic transport and deposition can sequester clay from coarser particles, they cannot efficiently separate clay-size kaolinite-metahalloysite, illite, smectite, anatase, goethite, hematite and organic matter. The so-called "sedimentary kaolins" originated not by sedimentational processes but, more particularly, by postdepositional alteration of sediments.

The original sediments were mainly neritic. They consisted of coarser micaceous and sometimes arkosic sands; impure kaolinitic and metahalloysitic fine sands and clays; and smectitic clays, marls and carbonates.

The Upper Cretaceous-Eocene muds that have been kaolinized were deposited in a dysoxic environment. The earliest postdepositional process to affect them was iron-stripping and sequestration, mediated by sulfate-reducing bacteria. These are very active at shallow depths below a sediment-seawater interface, where abundant sulfate is available and suitable organic matter is present. The HS^- generated during sulfate reduction reacts readily with Fe, including that in octahedral sites of kaolinite, to form FeS. Subsequent processes that affected these sediments were both abiotic and microbially mediated. In varying degrees, the kaolinizing processes destroyed smectite, illite/smectite and organic matter and leached alkalis, alkaline earths, silica, Fe and other metals. Where intensity and/or duration of kaolinization was sufficient, kaolins

formed. Diagenetic recrystallization and coarsening strongly modified most of the older kaolins and modified all of them to some degree.

Significantly, these kaolins are in a region where higher temperatures and rainfall long have induced more intense weathering and are restricted to areas where kaolinite-metahalloysite-rich detritus being transported to depositional sites was not diluted by harder-to-weather detritus from the continental interior. Both kaolins and associated bauxites are further restricted to the recharge area of a long-maintained regional groundwater system. The degree of postdepositional alteration relates closely to the pattern of groundwater movement and the boundaries of former or present unsaturated oxic zones.

The kaolinization that operated during the Cretaceous-Eocene interval continues to operate today. Its rate varied greatly from place to place, and commonly varied with time at the same place, in response to changes in imposed conditions. Kaolinization/bauxitization requires the influx of groundwater. In least permeable aquitards exposed only to dysoxic water, the rate of weathering was so slow as to produce only slight kaolinization in millions of years. In permeable layers or zones, however, where conditions were oxic and groundwater contained chelating organics or H_2SO_4 (from bacterially mediated oxidation of sulfides), the rate of weathering was high, and could accomplish strong kaolinization in a few thousand years.

Kaolins are the principal commercial product of postdepositional weathering in this region. Where total weathering was greater, gibbsitic kaolins developed. Where weathering was greater still, bauxite deposits formed, as in the Andersonville District of Georgia.

Implicit in use of the term "sedimentary kaolin" was a belief that they originated by sedimentary processes (Patterson and Murray 1984). Since accumulated data now show that they did not originate by sedimentary processes (Hurst 1997), calling them "sedimentary" is unjustified, even in allusion to their occurrence in sedimentary rocks, unless Cornwall kaolins (England) derived from granitic rocks are called "igneous" and Shaoping kaolins (China) derived from rhyolitic tuffs are called "volcanic".

The common practice of labeling a kaolin by the age of its host rocks ("Cretaceous kaolin" or "Eocene kaolin"), though convenient, is usually inaccurate because the kaolinization that produced it generally was not completed during its period of deposition.

When the evidence presented and referenced above for strong postdepositional kaolinization of earlier impure sedimentary rocks is accepted, the primary/secondary distinction sometimes used by clay scientists will become moot. As "primary" and "secondary" are defined in dictionaries and Bates and Jackson (1987), all kaolin minerals and all kaolins formed by weathering are secondary.

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