

developed countries limiting or eliminating production and release of many of these compounds, there are still concerns about the legacy of past releases to the environment present in coastal ocean surface sediments in several locations. There are serious coastal environmental and human health concerns associated with continued uses of several of these chlorinated hydrocarbons in developing countries.

See also

Crustacean Fisheries. Large Marine Ecosystems. Molluscan Fisheries. Network Analysis of Food Webs.

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CLAY MINERALOGY

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Introduction

Clay constitutes the most abundant and ubiquitous component of the main types of marine sediments deposited from outer shelf to deep sea environments. The clay minerals are conventionally comprised of the $< 2\ \mu\text{m}$ fraction, are sheet- or fiber-shaped, and adsorb various proportions of water.

This determines a high buoyancy and the ability for clay to be widely dispersed by marine currents, despite its propensity for forming aggregates and flocs. Clay minerals in the marine environments are dominated by illite, smectite, and kaolinite, three families whose chemical composition and crystalline status are highly variable. The marine clay associations may include various amounts and types of other species, namely chlorite and random mixed layers, but also vermiculite, palygorskite, sepiolite, talc, pyrophyllite, etc. The clay mineralogy of marine sediments is therefore very diverse according to depositional environments, from both qualitative and quantitative points of view.

As clay minerals are considered to be dependent on chemically concentrated environments, and as they commonly form in surficial conditions on land especially through weathering and soil-forming processes, their detrital versus authigenic origin in marine sediments has been widely debated. The transition from continental fresh to marine saline water, marked by a rapid increase of dissolved chemical elements, was the central point of discussion and arose from both American and European examples. In fact the mineralogical changes recorded at the land-to-sea transition are either important or insignificant, are characterized in estuarine sediments by various, sometimes opposite trends impeding consistent geochemical explanations, and often vanish in open marine sediments. The changes observed at the fresh-to-saline water transition in the clay mineral composition essentially proceed from differential settling processes or from mixing between different sources, and not from chemical exchanges affecting the crystalline network. Such a historical debate underlines the interest in investigating the sensitive clay mineral associations for understanding and reconstructing environmental conditions. This article will consider the general distribution and significance of clay minerals in recent sediments, some depositional and genetic environments, and a few examples of the use of clay assemblages to reconstruct paleoclimatic and other paleoenvironmental changes.

General Distribution and Significance

As a result of extensive reviews made by both American and Russian research teams the general

characters of the clay mineral distribution in deep sea sediments have been known since the late 1970s. The maps published by various authors demonstrate the dominant control of terrigenous sources, which comprise either soils and paleosoils or rocks. The impact of soils on the marine clay sedimentation is largely dependent on weathering intensity developing on land, and therefore on the climate. For instance, kaolinite mostly forms under intense warm, humid conditions characterizing the intertropical regions, and prevails in the clay fraction of corresponding marine sediments. By contrast chlorite and illite chiefly derive from physical weathering of crystalline and diagenetic sedimentary rocks outcropping widely in cold regions, and therefore occur abundantly in high latitude oceans. The kaolinite/chlorite ratio in marine sediments constitutes a reliable indicator of chemical hydrolysis versus physical processes in continental weathering profiles and therefore of climatic variations occurring on the land masses.

Other clay minerals are also able to bear a clear climatic message, as for instance the amount of random mixed layers and altered smectite in temperate regions, the crystalline status of illite in temperate to warm regions, and the abundance of soil-forming Al-Fe smectite in subarid regions. Detailed measurements on X-ray diffraction diagrams, electron microscope observations and geochemical analyses allow precise characterization of the different continental climatic environments from data obtained on detrital sedimentary clays.

Some terrigenous clay minerals in recent sediments reflect both climatic and non-climatic influences. For instance, the distribution of illite (Figure 1),

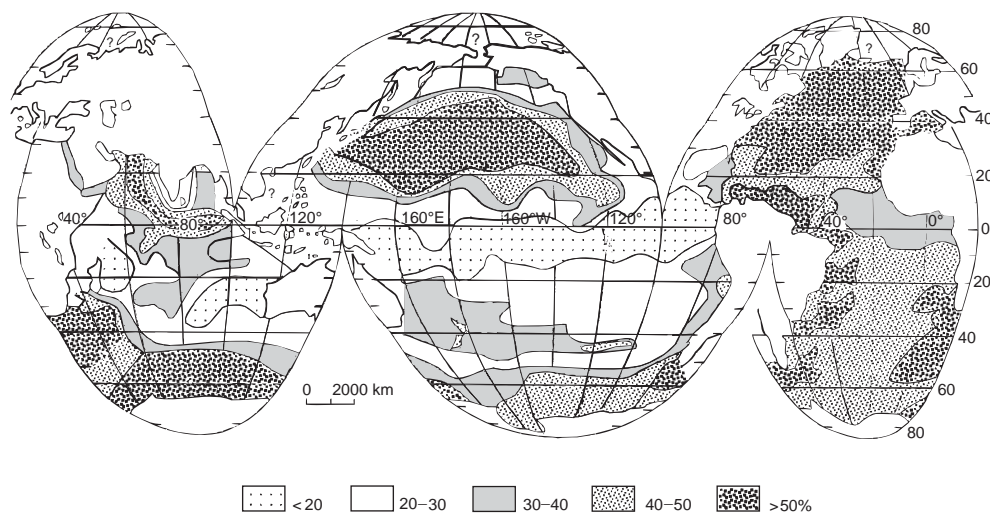


Figure 1 Worldwide distribution of illite in the clay fraction of surface sediments in the ocean. (After Windom, 1976. Reproduced with permission from Chamley, 1989.)

a mineral that primarily derives from the erosion of mica-bearing rocks, shows increased percentages in high latitude oceans due to predominant physical weathering, but also in a few low latitude regions depending on active erosion of tectonically rejuvenated, high altitude domains (e.g., supply by Indus and Ganges river drainage systems of Himalayan material to the northern Indian Ocean). The abundance of illite in the Atlantic Ocean, especially in its high latitude and northern parts, is due to several converging causes: cold to temperate climate, extensive outcrops of crystalline and metamorphic rocks, active erosion and river input, relative narrowness of the ocean favoring the ubiquitous transportation of the mineral particles, etc. Abundant illite percentages centered on the 30° parallel of latitude in the North Pacific result from aeolian supply by high altitude jet streams blowing from eastern Asia, and subsequent rainfall above the ocean. The general distribution of illite in marine sediments therefore proceeds from direct and indirect climatic control, meteorological conditions, petrographic and tectonic characteristics, physiography, river influx, etc.

All clay minerals may potentially be reworked from continental outcrops and transported over long distances until they settle on the ocean bottom. This is the case for nearly all geochemical types of smectite minerals (except perhaps for some very unstable ferriferous varieties formed in dense saline brines of the Red Sea), and also of palygorskite and sepiolite, two fibrous species wrongly suspected to not undergo significant transport. For instance, palygorskite and sepiolite are widely transported by wind and or water and deposited as detrital aggregates around the Tertiary basins bordering Africa and Arabia, where they initially formed under arid and evaporative conditions.

The clay mineral family whose distribution is the most complex and dependent on various detrital and autochthonous processes is the smectite group. Moderately crystalline smectites of diverse chemical types form pedogenically by chemical weathering under temperate conditions (essentially by degradation of illite and chlorite), and are supplied by erosion to sediments of mid-latitude regions where they are associated with various types and amounts of random mixed layers. Climate is also the dominant factor in warm, subarid regions where Al-Fe smectite forms in vertisolic soils and is reworked towards the ocean. Fairly high percentages of Fe-smectite characterize the low latitude eastern Pacific basins, where clay minerals in the clay-sized fraction are accessory relative to Fe and Mn oxides, and result from *in situ* hydrogenous genesis. In addition,

smectites of Fe, Mg, and even Al types may form by alteration of volcanic rocks, a process which is more intense in well drained, subaerial conditions (hydrolysis) than in submarine environments (halmyrolysis).

The diversity of the factors controlling the distribution of clay minerals in modern deep sea sediments is widely used to trace the influence of continental climate, geological and petrographic sources, tectonics, morphological barriers, etc., and also to identify the nature, direction and intensity of transportation agents. As an example, the distribution of smectite and illite in the western Indian Ocean depends on different source provinces as well as on land geology, climate, volcanism, aeolian and marine currents (Figure 2). The terrigenous sources and climatic conditions relieved by north-to-south or south-to-north surface to deep currents are responsible for long-distance transportation of Antarctic-derived smectite in the Crozet and Madagascar basins, of abundant volcanogenic smectite derived from Deccan traps erosion off the Indian coasts, of Himalayan illite in the Indus and Ganges deep sea fans, of illite associated with up to 30% palygorskite off Arabian and especially on submarine ridges (i.e., aeolian supply), and of illite associated with soil-derived kaolinite off Southeastern Africa. Both illite and smectite are dominantly inherited from various terrestrial rocks and soils, including Antarctic outcrops responsible for illite dominance to the west of the Indian Ocean (35°C) and for smectite dominance to the east (45–75°C). An *in situ* smectite-rich province located in the southern ocean around 55°S and 70°E is attributed to the submarine alteration of volcanic rocks. Volcanic contributions are also suspected in the Central Indian basin and in the vicinity of Indonesia. Of course such investigations constitute very useful guidelines for reconstructing past climatic, oceanographic, and physiographic conditions.

Marine Autochthonous Processes

From Volcanic to Hydrothermal and Hydrogenous Environments

Until the 1970s, the submarine weathering of volcanic material (basalt, glass, ash) was often considered to be responsible for important *in situ* formation of clay minerals, especially of smectite, in deep sea sediments. Effectively basalt altered by surficial oxidation and hydration may give way to Mg-smectite, sometimes Fe-smectite, frequently associated with celadonite (a glauconite-like Fe-Al micaceous species), phillipsite (a Na-rich zeolite),

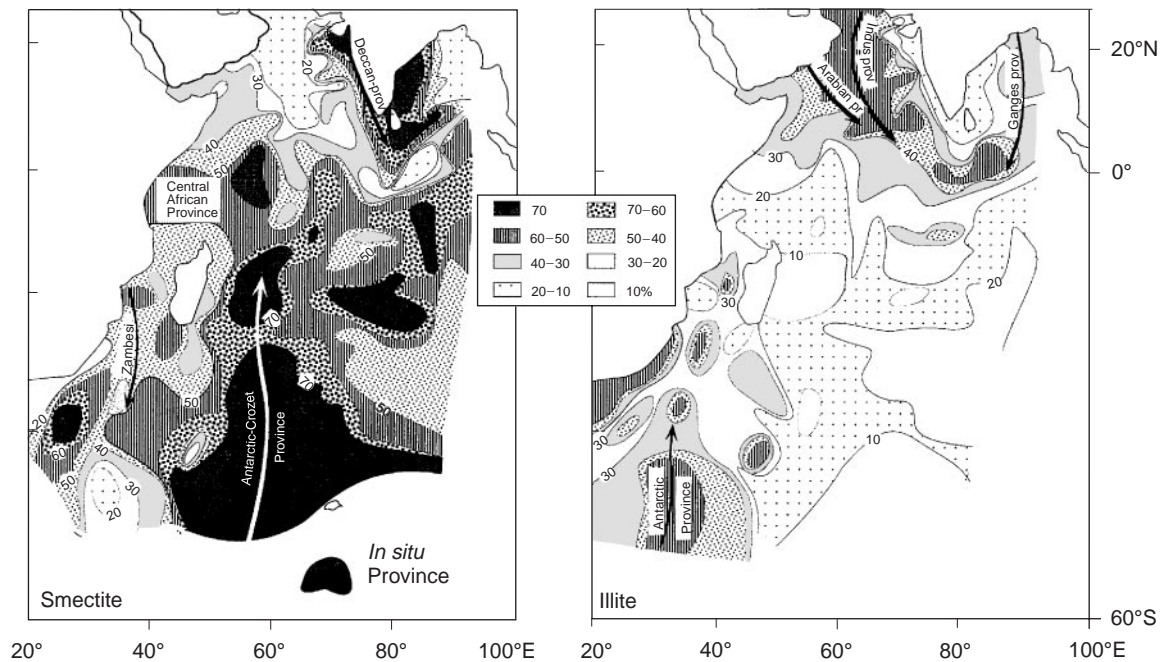


Figure 2 Distribution of smectite and illite in the western Indian Ocean, and related source provinces. (After Kolla *et al.*, 1976. Reproduced with permission from Chamley, 1989.)

calcium carbonates, Fe-Mn oxyhydroxides, etc. The more amorphous, the smaller sized and the more porous the volcanic material (e.g., pumiceous ashes), the more intense the submarine formation of clay. In fact the clay minerals resulting from halmyrolysis of volcanic material are quantitatively limited and essentially located at close vicinity to this material (e.g., altered volcanoclastites or basalts); they are unable to participate in a large way in the formation of the huge amounts of clay incorporated in deep-sea sediments. Additional arguments contradicting the importance of volcanic contribution to deep-sea clay consist of the frequent absence of correlation between the presence of volcanic remains and that of smectite, and in the non-volcanogenic chemistry of most marine smectites (e.g., aluminum content, rare earth elements, strontium isotopes). The shape of smectite particles observed by electron microscopy is typical of volcanic influence only in restricted regions marked by high volcanic activity, especially explosive activity. Notice that local overgrowths of lath systems oriented at 60° from each other may characterize marine clay particles and especially smectites, but they are neither specifically related to volcanic environments nor associated with noticeable increase of smectite proportion or specific change in the clay chemical or isotopic composition. The intrusion of basalt sills in soft marine sediments may determine some metamorphic effects and the very local formation of

ordered mixed layers (corrensite), chlorite, and associated non-clay minerals.

The hydrothermal impact on deep-sea sedimentation is fundamentally characterized by *in situ* precipitation of Fe-Mn oxyhydroxides relatively depleted in accessory transition elements (Co, Cu, Ni), and locally by the deposition of massive sulfides near the vents where hot and chemically concentrated water merges. The autochthonous clay minerals in such environments are marked by various species depending on fluid temperature, oxidation-reduction processes, and fluid/rock ratio. For instance, drilling holes in Pacific hydrothermal systems show different mineral evolutions. In the hydrothermal mounds of the Galapagos spreading center, the fluids are rich in silicon and iron and of a low temperature (20° – 30°C) throughout the 30 m-thick sedimentary column; this gives way in oxidized conditions to the precipitation of Fe-smectite as greenish layers interbedded in biogenic oozes that at depth evolve into glauconite by addition of potassium (Figure 3A). By contrast the detrital to authigenic deposits of the Middle Valley of Juan de Fuca ridge show on a 40 m-thick series the *in situ* formation from high temperature Mg-rich fluids (200°C) of a downwards sequence characterized by saponite (a Mg-smectite), corrensite (a regular chlorite-smectite mixed layer), swelling chlorite, and chlorite (Figure 3B). At this site geochemical and isotope investigations reflect a noticeable downhole

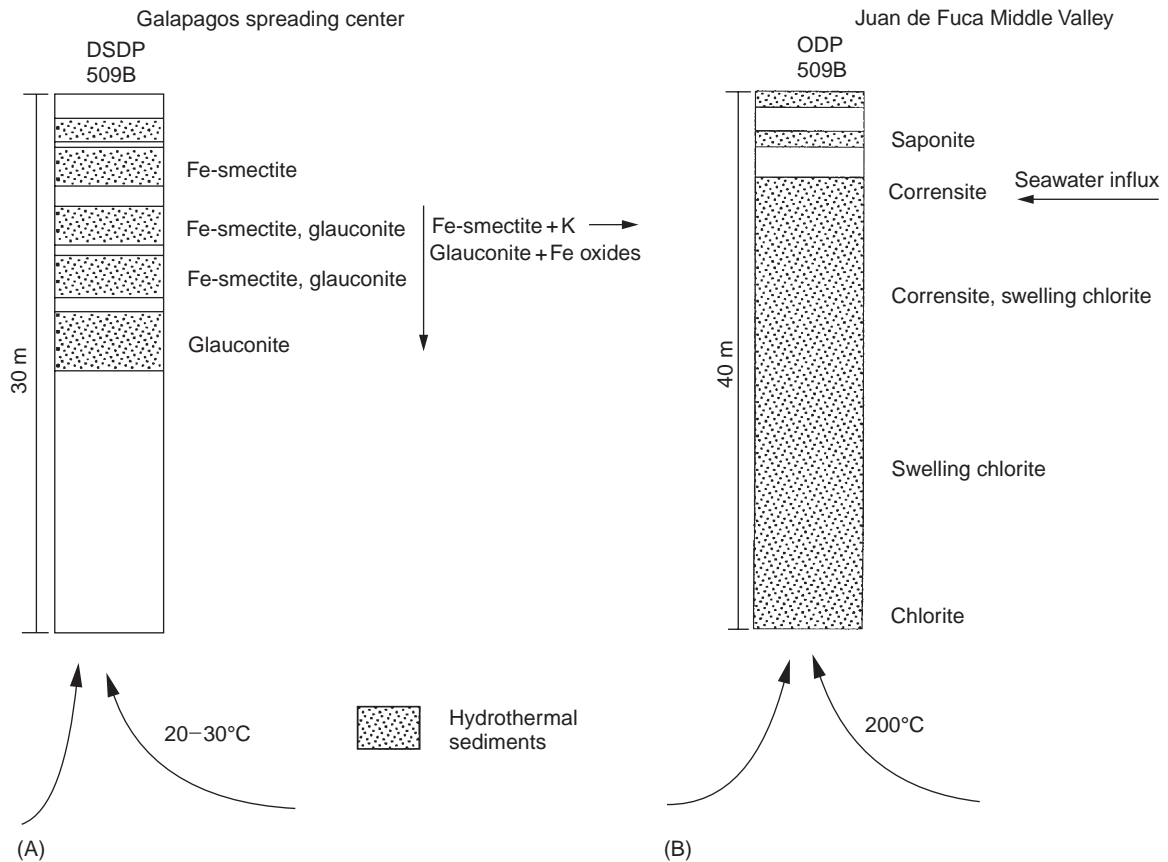


Figure 3 Schematic vertical distribution of typically hydrothermal clay minerals in the sedimentary systems of (A) the Galapagos spreading center, and (B) the Juan de Fuca Middle Valley. (Reproduced with permission from Buatier and Karpoff, 1995.)

increase of temperature and strong changes in the fluid composition.

A more widespread process consists of the **hydrogenous** formation of clay at the sediment–seawater interface, in deep-sea environments characterized by water depths > 4000 m, insignificant terrigenous supply, and very low sedimentation rate (< 1 mm/1000 years). This is particularly the case for some Central and South Pacific basins. The sediments mostly consist of reddish-brown oozes rich in Fe and Mn oxides (i.e., ‘deep sea red clay’). There iron-rich smectites of the nontronite group may form in significant proportions, probably due to long-term low temperature interactions between (1) metal oxyhydroxides supplying the iron, (2) seawater supplying the magnesium and other minor to trace elements, (3) biogenic silica supplying the silicon, and (4) allochthonous accessory particles (e.g., aeolian clay) supplying the other chemical elements (e.g., Al). Notice that the distinction between pure hydrothermal and pure hydrogenous clay minerals forming on the deep-sea floor necessitates detailed chemical analyses (Table 1) and often additional microprobe and isotope investigations.

To summarize, the distribution of clay minerals in deep sea deposits marked by active volcanic-hydrothermal activity and by very low sedimentation rates depends on various and complex *in situ* influences among which the hydrogenous processes quantitatively prevail. The distinction of these autochthonous influences is complicated both in the vicinity of land masses where terrigenous supply becomes active, and in shallower areas where biogenic influences may intervene more intensely (e.g., Nazca plate, south-east Pacific).

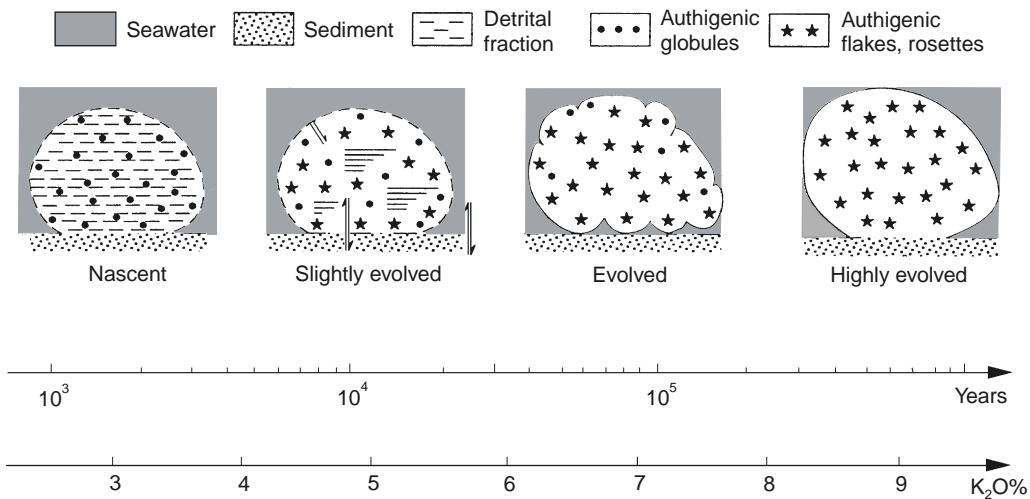
Ferrous Clay Granules

Iron-rich clay granules are traditionally called glauconite, which is somewhat incorrect as glauconite is a specific clay mineral, whereas clay granules may include various iron-bearing clay species. Ferriferous clay granules form on continental margins at water depths not exceeding 1000 m, and comprise two major types characterized by specific colors, clay minerals, and habits. Glaucony, the most widespread type, constitutes dark green to brown clayey aggregates, and may comprise different varieties of iron-rich illite- and smectite-like minerals such as

Table 1 Examples of chemical composition of hydrothermal to hydrogenous smectites in Central and South Pacific sediments

Type of smectite	Tetrahedra			Octahedra			Interlayers		
	Si	Al	Fe	Al	Fe	Mg	Ca	NH ₄	K
Pure hydrothermal (Galapagos mounds field)	3.94	0.06	—	0.03	1.59	0.38	0.03	—	0.36
Hydrothermal and hydrogenous (Bauer Deep)	3.97	0.03	—	0.44	1.07	0.54	0.05	—	0.06
Hydrogenous > hydrothermal (Galapagos spreading centre)	3.97	0.03	—	1.12	0.48	0.37	0.09	—	0.11
Pure hydrogenous (?) (North Marqueses fracture zone)	3.37	0.63	—	0.39	1.12	0.46	0.46	—	0.17

(Reproduced with permission from Chamley, 1989.)

**Figure 4** Successive stages of glaucony formation from a pre-existing substrate. (Reproduced with permission from Odin, 1998.)

glaucinite (Fe- and K-rich illitic clay), Fe-smectite, and Fe illite-smectite mixed layers. Glaucony may form at latitudes as high as 50° and in water depths as great as 1000 m, but usually occurs in 150–300 m water depths at the shelf-slope transition of temperate-warm to equatorial regions. Verdine, which is less ubiquitous and has been identified more recently, constitutes light green to light brown granules characterized by phyllite V or odinite, a ferriferous clay mineral of the kaolinite family (described by G.S. Odin, who has developed outstanding investigations on clay granules). Verdine forms in rather shallow water sediments (maximum 50–80 m) of intertropical regions, and depends on the supply of abundant dissolved iron by low latitude rivers.

Ferriferous clay granules form at the sediment–water interface and evolve at burial depths rarely exceeding a few decimeters. They develop in semi-confined environments at the expense of various substrates submitted to ‘greening’: chiefly fecal

pellets and microfossil chambers (e.g., foraminifera), calcareous or siliceous bioclasts, minerals (especially micas), and rock debris. The formation of glaucony (which somewhat leads to diffuse habits), occurs in successive stages marked by a rapid and strong enrichment of iron and then potassium, a volume increase causing external cracks, and the obliteration of the initial shape (Figure 4). The formation of verdine still has to be documented, but both clay granule types correspond to true authigenic formation rather than to transformation of pre-existing clay minerals. The chemical evolution of ferriferous clay granules vanishes either after a long exposure at the sediment–water interface (10^5 – 10^6 years for glaucony), or after significant burying.

Organic Environments

The influence of living organisms on clay-rich sediments is mainly marked by physical processes referred to as bioturbation, and concerns various

marine environments, especially on continental shelves. Chemical modifications of clay associations are only occasionally reported and seem to affect the crystalline status of chlorite and associated random mixed layer clays locally through ingestion and digestion processes of shallow water crustaceans, annelids or copepods. The chemical interactions developing in digestive tracts between clay minerals and organic acids appear to have small quantitative effects, as the marine clay associations are roughly the same as the terrestrial associations.

The chemical impact on clay mineral stability of the organic matter incorporated in deep marine sediments is variable. Most sedimentary series containing significant amounts of dispersed organic matter (i.e., 1–3%) do not display any specific clay mineral composition. For example, this is the case for black shales deposited during the Cretaceous period in the Atlantic, where clay mineral associations may comprise vulnerable species such as smectite and palygorskite, the abundance and crystalline status of which vary independently of the content and distribution of the organic matter. In contrast, the sapropels developing in the eastern Mediterranean during the late Cenozoic era, especially in Quaternary high sea level stages, show some *in situ* degradation processes of the detrital clay minerals (Figure 5). Submarine alteration af-

fects the mineral species in successive stages depending on their ability to resist acid conditions. Palygorskite is the more vulnerable species and kaolinite the more resistant. The degradation of clay assemblages tends to increase toward the central and deepest parts of marine basins, in depressed morphological zones, and at the base of the decimeter-to-meter thick sapropels. The degradation of clay minerals under organic conditions has occurred close to the sediment–water interface and appears to depend on the chemical nature and evolution stage of the terrestrial and marine organic matter.

Paleoenvironmental Expression

Clay mineral assemblages of sediments successively deposited in marine basins express various environmental messages related to the geological history. A few examples from recent Quaternary to late Cenozoic series will be considered here. Similar messages may be preserved in much older series of Mesozoic and even Paleozoic ages, provided that the diagenetic imprint due to lithostatic overburden, geothermal gradient, and fluid circulation has remained moderate. Clay-rich, low permeability sedimentary formations 2–3 km thick and submitted to normal heat flow (c. 30°C/km) are usually prone to preserve such paleoenvironmental characteristics.

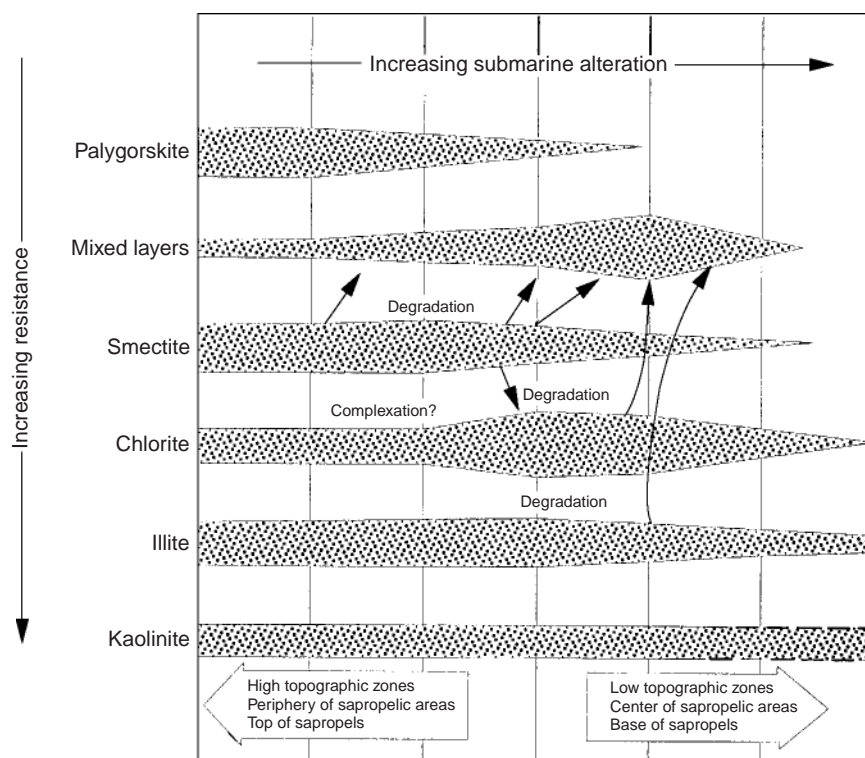


Figure 5 Characters of the clay mineral degradation in Quaternary sapropels of the eastern Mediterranean basins.

Climate

As clay minerals at the surface of the Earth are dominantly formed through pedogenic processes depending on climate and are particularly subjected to surficial erosion and reworking, their assemblages successively deposited in a given sedimentary basin are *a priori* able to reflect successive climatic conditions that prevailed on adjacent land masses. This implies that very little post-depositional, i.e., diagenetic changes have affected the clay assemblages after their storage in sediments. This is observed to be the case in many series drilled or cored in the oceans. The climatic message borne by clay has been documented by numerous investigations, and corroborated by the comparable range of variations recorded in the nature and proportions of clay minerals in both present-day soils outcropping at various latitudes and marine sedimentary columns. Marine clay mineral assemblages basically express the type and intensity of continental weathering, which depend predominantly on the ion leaching through the action of humidity and temperature, and secondarily on seasonal rainfall and drainage conditions.

Quaternary glacial-interglacial alternations caused terrestrial alternation of physical and chem-

ical weathering processes, and this was reflected in the clay assemblages successively brought to marine sediments through soil erosion and river or wind transport. Sedimentary levels contemporary with cold periods are usually characterized by more abundant rock-derived minerals such as richly crystalline illite, chlorite, smectite and associated feldspars reworked from active physical weathering. Warm, humid periods generally correspond to increased supply of soil-derived kaolinite and metal oxides, poorly crystalline smectite and various random mixed layer clay minerals. For instance, the terrigenous fraction of hemipelagic sediments deposited from 500 000 to 100 000 years ago in the Northwestern Atlantic off New Jersey and dominantly derived from the erosion of Appalachian highlands shows increased proportions of chlorite in glacial isotopic stages, and of kaolinite in interglacial stages. This is clearly expressed by the kaolinite/chlorite ratio (Figure 6). Paleoclimatic reconstructions from clay mineral data are available for various geological periods, as for instance the passage since about 40 Ma from a non-glacial world dominated by chemical weathering (smectite, kaolinite) to a glacial world in which physical weathering was greater (chlorite, illite). The comparison of climatic curves provided by clay minerals

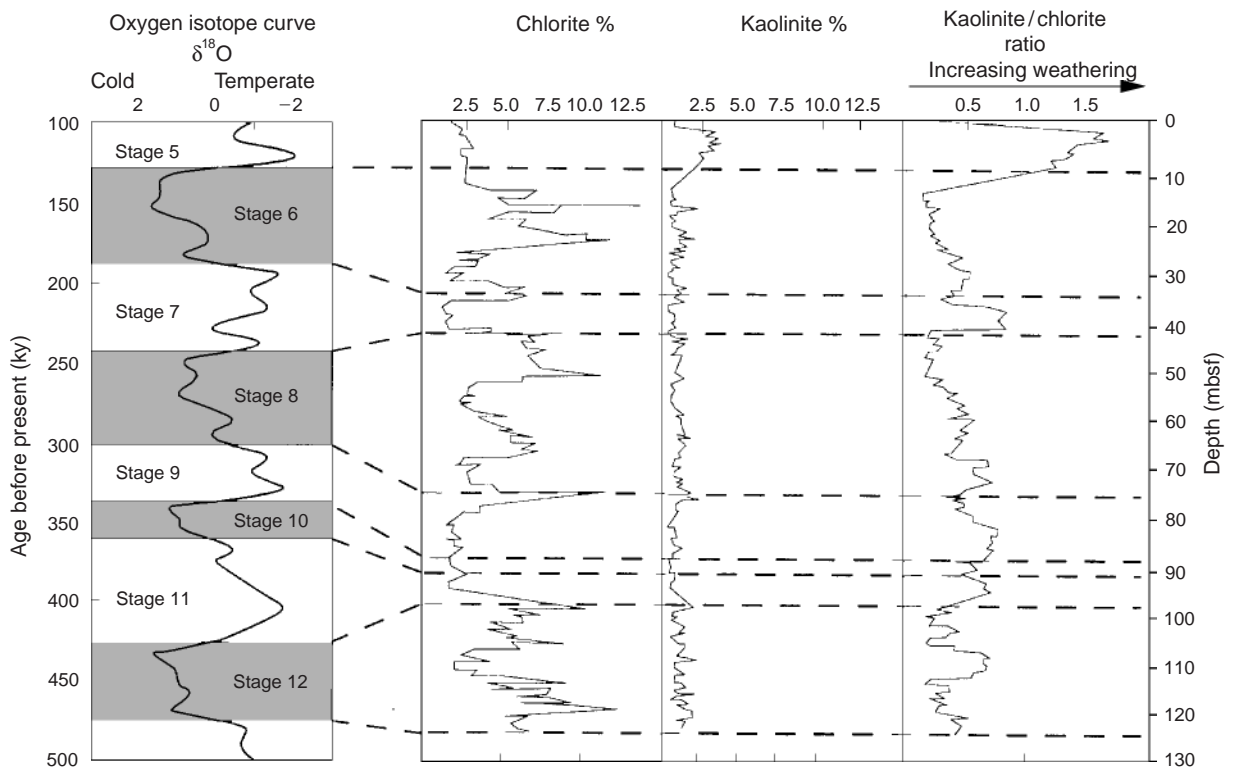


Figure 6 Comparison and climatic significance of clay mineral and oxygen isotope data from stages 12 to 5 at ODP Site 902, New Jersey continental margin. (Reproduced with permission from Vanderaveret *et al.*, 1999.)

and other indicators (oxygen isotopes, micro-faunas or -floras, magnetic susceptibility, etc.) allows a better understanding of the nature, intensity, and effect of the different factors characterizing the terrestrial and marine climate in given regions during given geological intervals.

High resolution studies show that clay assemblages may express terrestrial climatic variations at a centennial scale or even less, and that the influence of Earth's orbital parameters varies to different extents according to the latitude. For example, the clay minerals data of Quaternary North Atlantic deep sea sediments were submitted to cross-correlation spectral analyses on 5.5–14 m-long cores encompassing the last 300 000 years. The mineral composition displays a general 100 000-year cyclic signal (eccentricity) in the whole 45°–60°N range, a 41 000-year signal (obliquity) at highest latitudes related to dominant aeolian supply, and a 23 000-year signal (precession) at mid-latitudes related to dominant transport by marine currents (Table 2).

The paleoclimatic expression by clay mineral successions is direct or indirect, i.e., it either indicates the climate that actually prevailed at a given period, or reflects other events depending on climate: migration of lithospheric plates across successive climatic zones, varying extension of ice caps controlling the surficial erosion, variations in the marine circulation regime due to changing latitudinal and vertical heat transfers. The direct paleoclimatic reconstructions from clay mineral data are all the more reliable since the marine basins investigated are preserved from important erosion of paleosoils, changes in detrital sources, differential settling processes, longitudinal oceanic currents, and major geomorphological changes.

Marine Currents

The different marine water masses may carry the small and light clay mineral particles over long distances, and therefore leave an imprint within the sediments at the depth range they are moving. This has been demonstrated for late Quaternary sediments of the southwestern Atlantic, where the southward-flowing North Atlantic deep water is enriched in kaolinite supplied from rivers draining the intertropical South American continent, and the northward flowing Antarctic Atlantic bottom water supplies chlorite and smectite issuing from southernmost Argentina and Antarctica. Paleocurrent reconstructions from clay data exist mainly about Atlantic and Southern Oceans, which are marked by numerous and distinct terrigenous sources, vertical mixing and longitudinal heat transfers, and Tertiary to Quaternary changing conditions of the superimposed water masses volume and celerity.

Tectonic Activity

The tectonic instability determines some changes in the composition of clay mineral assemblages which are usually much more important than those due to climate or circulation. First, the subpermanent rejuvenation by neotectonics of continental relief increases the erosion potential and therefore impedes the development of pedogenic blankets where clay minerals tend to be in equilibrium with current climatic conditions. Such a chronic tectonic activity explains the abundance of rock-derived illite and chlorite in equatorial Indian Ocean basins depending on Himalayan output. Second, a continental tectonic uplift determines changes in the nature of clay minerals eroded from rocky substrates, while

Table 2 General relationships between the clay mineral distribution and the three main Earth's orbital frequency bands according to latitude, from cross-correlation spectral analysis of X-ray diffraction data on North Atlantic cores

Core	SU 90-08			SU 90-12			SU 90-38			SU 90-33		
	44°N			51°N			54°N			60°N		
Orbital parameters	E	O	P	E	O	P	E	O	P	E	O	P
Illite	H	—	V	H	—	V	V	H	—	H	V	—
Chlorite	V	—	V	V	—	V	V	V	—	V	V	—
Kaolinite	H	—	V	H	—	—	V	V	—	H	V	—
Illite-vermiculite	—	—	—	—	—	V	—	—	—	—	—	—
random mixed layer												

E, eccentricity band, 100 000 year; O, obliquity band, 41 000 year; P, precession band, 23 000 year; H, high variance power; V, very high variance power. Maximum correlations in bold characters. (Reproduced with permission from Bout-Roumzeilles *et al.*, 1997.)

submarine uplift may determine morphological barriers to the clay transfer. This was the case for the Hellenic Trench in the eastern Mediterranean during late Pliocene to early Pleistocene periods, when the combined uplift of Peloponnese and of Mediterranean ridge both increased the terrigenous input of European illite and chlorite and blocked the supply of African palygorskite. Due to their sensitivity to geomorphological changes and their aptitude for long distance transportation, clay minerals are able to express slight and progressive epeirogenic changes as well as very remote tectonic events.

Sea also

Aeolian Inputs. Cenozoic Climate – Oxygen Isotope Evidence. Deep Sea Drilling Results. Hydrothermal Vent Deposits. Rare Earth Elements and their Isotopes in the Ocean. River Inputs. Water Types and Water Masses.

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CLIMATE

See **FISHERIES AND CLIMATE; METHANE HYDRATES AND CLIMATIC EFFECTS; PLANKTON AND CLIMATE**

CLIMATE CHANGE

See **ABRUPT CLIMATE CHANGE; HOLOCENE CLIMATE VARIABILITY; MILLENIAL SCALE CLIMATE VARIABILITY; SEABIRD RESPONSES TO CLIMATE CHANGE**

CLIMATE MODELS

See **EL NIÑO SOUTHERN OSCILLATION (ENSO) MODELS; PALEOCEANOGRAPHY, CLIMATE MODELS IN**