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Abstract

Maps of the distributions of the four major clay minerals (smectite, illite, kaolinite and chlorite) in and around the Mississippi River drainage basin and in the Northern Gulf of Mexico have been produced using newly acquired data from erodible/alluvial terrestrial sediments and marine surface sediments, as well as from previously published data. East of the Rockies, North America can be divided into four, large, clay-mineral provinces: (1) the north-western Mississippi River watershed (smectite rich), (2) the Great Lakes area and eastern Mississippi River watershed (illite and chlorite rich), (3) the south-eastern United States (kaolinite rich) and (4) the Brazos River and south-western Mississippi River watersheds (illite and kaolinite rich). The clay mineral distributions in surface sediments of the present-day Gulf of Mexico are strongly influenced by three main factors: (1) by relative fluvial contributions: the Mississippi River delivers the bulk of the clay input to the Northern Gulf of Mexico whereas the Apalachicola, Mobile, Brazos and Rio Grande rivers inputs have more local influences; (2) by differential settling of various clay mineral species, which is identified for the first time in Northern Gulf of Mexico sediments; and (3) by oceanic current transport: the Gulf of Mexico surface and subsurface circulation distributes the clay-rich sediments from river mouth sources throughout the Northern Gulf of Mexico.

1. Introduction

Clay minerals are a dominant component of most marine sediments, and are mainly land-derived (Biscaye, 1965; Griffin et al., 1968). Their geographic distributions and sources

have been extensively investigated since the 1960s (Biscaye, 1965; Griffin and Goldberg,1969; Rateev et al.,1969; Gradusov,1974; Windom,1976). Studies of clay mineral assemblages provide information on climatic conditions, such as precipitation and runoff patterns over the adjacent continents (Chamley,1989), as well as on the dynamics of river inputs (Pinsak and Murray, 1960; Scafe and Kunze, 1971; Doyle and Sparks, 1980). Clay minerals can be advected over long distances and then settle far away from their source area, especially if being re-transported in the nepheloid layer (Biscaye and Eittreim, 1977; Jones, 1984). Thus, clay mineral assemblages have also been successfully used to trace oceanic current patterns (Petschick et al., 1996; Fagel et al., 1997; Gingele et al., 2001; Boulay et al., 2005).

The Gulf of Mexico has been an important area for global thermohaline circulation and for the global climatic system (Broecker, 1991) especially since the closure of the Isthmus of Panama 4.6 million years ago (Haug and Tiedemann, 1998). Gulf of Mexico surface waters are characterized by high temperature and high salinity that further influence the Gulf Streamwater properties. The hydrological features of the Gulf of Mexico were strongly modified by the inputs of freshwater resulting from melting pulses of the Laurentide Ice Sheet (LIS) during the last glacial cycle and the last deglaciation (Kennett and Shackleton, 1975; Leventer et al., 1982; Joyce et al., 1993; Aharon, 2003; Flower et al., 2004). Changes of salinity resulting from these melting pulses are thought to have altered the thermohaline circulation, the global heat exchange and worldwide climatic conditions (Broecker et al., 1990; Bond, 1995; Rahmstorf, 1995; Manabe and Stouffer, 1997). Although the direct palaeoceanographic effects of these freshwater supplies on the Gulf of Mexico have been studied largely using d18O variations, the associated terrigenous inputs (mostly clay minerals; Brown and Kennett, 1998) have been poorly investigated. They may, however, provide information on the varying continental provenance of these freshwater pulses and therefore help close the gap between climate variations over the continent (e.g. the dynamics of the LIS/ proglacial lake overflows and/or increase in precipitation; Broecker et al., 1989; Teller, 1995; Clark et al., 2001; Clarke et al., 2003) and the hydrological changes in the Gulf of Mexico. In order to use clay mineral records from the Gulf of Mexico for palaeoclimate reconstructions (Sionneau et al., in preparation), one must have a good knowledge of clay mineral continental sources and of the factors influencing their distribution in the Gulf of Mexico. It seems reasonable to assume that the nature of the main clay mineral provinces in North America has remained unchanged during the late Quaternary. Variations in clay mineral assemblages in the Gulf of Mexico surface sediments are then mainly controlled by

changes in erosion and transport pattern, even if their intrinsic nature derived from weathering processes developed over longer timescale (Thiry et al., 1999; Thiry, 2000).

The present Gulf of Mexico circulation pattern is relatively well known (Fig. 1). Its sedimentology has been extensively studied (Griffin, 1962; Doyle and Sparks, 1980; Balsam and Beeson, 2003; Ellwood et al., 2006). None of these studies, however, combined data from both continental and marine environments, and no overall clay mineral distributions over North America and the Northern Gulf of Mexico are available today.

In this paper, we present the clay mineral assemblage of 28 continental sites [consisting of 85 new samples and data from Griffin (1962) and Potter et al. (1975)] over North America and of 67 core-top samples collected over the Northern Gulf of Mexico. The main objectives of this study are: (1) to map clay mineral distributions over both the North American continent east of the Rockies and the Northern Gulf of Mexico; (2) to discern and distinguish the continental sources of the principal marine clay minerals; and (3) to identify the main parameters (fluvial contributions, differential settling, oceanic currents) controlling the present-day marine distribution of these clay minerals in the Northern Gulf of Mexico.

2. Samples

Marine core-top samples were recovered during the RVs Robert Conrad (RC), Vema (VM) and Atlantis (AT) cruises (Lamont core collection; Table 1; samples 13–67) as well as during the RV Marion Dufresne IMAGES cruise PAGE in 2002 (Bout-Roumazeilles and Trentesaux, 2006; samples 3–12) and PICASSO in 2003 (Laj et al., 2004; samples 1 and 2). They are broadly distributed over the northern Gulf of Mexico (Fig. 2) and are located within the different geomorphologic provinces that characterize this area (Fig. 3).

Over the North American continent, 85 samples were collected from different types of sediment (loess, recent mineral dust deposits, tills, lake and river) at 14 sites (Fig. 4, Table 2). Sediment samples of fluvial origin were taken from the C horizon of alluvial plain soils (about 1.5–2 m depth) from which, except for the carbonate content, the mineralogical composition is not likely to have been disturbed by anthropogenic activity and should reflect the bulk composition of superficial deposits such as loess, alluvium or tills (Gallet et al., 1998). All these continental samples are commonly representative of the regional and/or upstream watershed average mineralogical composition. In addition we have included published clay mineral data from 11 sites on the Mississippi river system (Potter et al., 1975) and from the Mobile Embayment, the Mississippi and the Apalachicola River mouths (Fig. 5, Table 2; Griffin, 1962). The majority of the continental sites (20 of 28) are located in the

present-day Mississippi River watershed (Fig. 4).

3. Methods

X-ray diffraction analyses (XRD) were performed at the University of Lille and the LDEO (Lamont–Doherty Earth Observatory of Columbia University). In this paper, ‘‘clay minerals’’ refer to the major phyllocilicate minerals within the clay-size fraction (generally <2 mm). They are identified by XRD following the protocol of Holtzapffel (1985). All samples were first decalcified with 0.2 N HCl. The excess acid was removed by repeated washings with distilled water and centrifugations. The clay-size fraction was separated by settling according to Stokes's law, concentrated by centrifugation, and oriented by wet smearing on glass slides. The analyses were run on a Philips PW 1749 generator (copper anode; 40 kV voltage; 25 mA intensity and 1°2q/min speed) at the University of Lille, and on a Philips X'Pert-MPD (q–q goniometer; Cu X-ray tube; Peltier-cooled, high sensitivity, Kevex energy dispersive detector) at the LDEO. A complete X-ray diffraction analysis requires several runs to permit precise mineral identification. Two XRD runs were performed routinely for each sample: (1) air-dried sample (‘‘normal’’ run) and (2) ethylene-glycol vapour saturation for 12 h (‘‘glycol’’ run). The analyses were run from 2.49° to 32.49° 2θ. Each clay mineral is characterized by its basal layer plus interlayer interval (d) as revealed by XRD analysis (Brown and Brindley, 1980). Smectite (‘‘S’’) is characterized by a peak at 14 Å on the normal run, which expands to 17 Å after saturation by ethylene glycol. Illite (‘‘I’’) presents a basal peak at 10 Å on the two runs. Chlorite (‘‘C’’) is characterized by peaks at 14 Å (001), 7 Å (002), 4.75 Å (003) and 3.53 Å (004) on the two diffraction spectra. Kaolinite (‘‘K’’) is characterized by peaks at 7 Å (001) and 3.58 Å (002) on the normal and glycol runs. To distinguish kaolinite from chlorite, the portion of the spectrum containing the basal peaks of kaolinite (002) and chlorite (004) around 3.55 Å is step-scanned in a high-resolution mode following standard procedures described in detail by Petschick et al. (1996). Semi-quantitative estimation of clay mineral abundances is based on peak areas, weighted by empirically estimated factors (Biscaye, 1965; Biscaye et al., 1997) and summed to 100% (S + I + K + C = 100%). Peak area measurements were realized in the glycol runs using the Macintosh Mac- Diff® 4.2.5 software (Petschick, 2000). Results are given in Tables 1 and 2. The error on the reproducibility of measurements is estimated to be ±5% for each clay mineral. The mixed-layered clay minerals identified by Potter et al. (1975) in Ohio River and Tennessee River samples (sites 14 and 15) were not taken into account in the percentages since their exact nature (vermiculite and/or illite–chlorite and/or illite–smectite) is unknown.

All data were recalculated according to the S + I + K + C = 100% formula in order to be compared to each other. The data have been gridded and contoured using the Surfer® 8 software. For each clay mineral, scales between isolines of land and sea maps are different to allow a better representation of their relative variations in each environment. Map contours do not represent precise boundaries of clay mineral composition on land, but rather indicate rough limits between the dominant source areas of each clay mineral over the United States, based on our dataset.

4. Clay mineral distribution

In most continental environments, illite and chlorite are usually inherited from ancient rocks (Palaeozoic, pre-Cambrian) modified by physical or moderate chemical weathering at high latitudes or altitude where Quaternary periglacial climatic conditions prevail. In North America, kaolinite forms through long-term weathering processes during preglacial time (Thiry et al., 1999; Thiry, 2000). It can derive from saprolites as well as from old sedimentary formations and may also form in solfatares in association with smectite (Huertas et al., 1999). Smectite is either derived from volcanic ash layers or authigenic in poorly-drained environments as in alluvial or coastal plains (Kraus, 1999; synthesis in Gerrard et al., 2007). Smectite can also be locally formed in the marine environment through early diagenesis, halmyrolysis (Karpoff et al., 1989), or hydrothermal weathering of volcanic rocks (Chamley, 1998).

4.1. Clay mineral distributions in and around the Mississippi River watershed

Three large areas can be distinguished based on smectite abundance patterns (Fig. 6a). The first covers a huge part of the Mississippi River watershed, including the Missouri River basin and Mississippi River alluvial plains. A corridor of high smectite values (>40%) extends from Saskatchewan to the Mississippi River mouth and separates two areas of low smectite content (<20%): the eastern Mississippi River and the north-western Texas watersheds. Smectite decreases rapidly (from 45% to 0% in few hundreds of kilometres) from the Mississippi River toward the eastern part of the United States of America (USA).

Due to the calculation method used, clay mineral percentages are not independent of each other. Because clay minerals are summed to 100%, variations in the two most abundant ones (smectite and illite) have opposite trends. Two major zones characterized by high illite concentrations can be observed in the distribution pattern of that mineral (Fig. 6b). Sediments exposed near the Great Lakes area are characterized by illite values >40%, especially the

areas northeast and southwest of Lake Huron. Similarly the clay mineral fraction of sediments from north-western Texas (Gulf Coast Rivers of the south-western USA and southwestern Mississippi River watersheds) contain more than 40% illite. Those two zones are separated by an illite depletion extending from the north-western Mississippi River basin to the south-eastern USA. This zone is composed of three minimal areas (<15%): Saskatchewan, the lower Arkansas River and Alabama-Florida coastal plains between the Mobile Embayment and the Apalachicola River mouth.

The south-eastern USA where soils and sediments are very rich in kaolinite (>30%) is one of the most important sources of this mineral. Kaolinite proportions are maximal in north Florida, exceeding 60% (Fig. 7a). Two other potential source areas for kaolinite, characterized by values around 20%, are located in the upper Mississippi River and around the Pecos River in Texas. The Missouri River system and the middle and lower Mississippi River alluvial plain sediments display low kaolinite content, contrasting with the high values to the west and to the east.

The chlorite distribution displays an East-West decreasing gradient (Fig. 7b). High values of chlorite (>20%) are found in the eastern part of the USA (east of about 087° W). Chlorite abundance decreases westward, first sharply down to about 10%, in the Mississippi River valley, and then more gradually, down to <4%, toward the Gulf of Mexico coastal plains, the western Mississippi River watershed and north-western Texas.

Since in most continental environments, smectite and kaolinite are generally formed by chemical weathering, while illite and chlorite are mostly inherited from ancient rocks modified by physical or moderate chemical weathering, the clay mineral ratio (smectite + kaolinite)/(illite + chlorite) indicates the type of weathering (chemical or physical) that affected the erodible sediments. According to the distribution of this (S + K)/(I + C) ratio (Fig. 8), the North American continent east of the Rockies can be divided in different dominant weathering-regime areas.

The south-eastern United States is characterized by maximum values of the ratio (>4), suggesting that chemical weathering prevails in this region. This seems to be in agreement with the presence of yellow-red subtropical soils (Fig. 5). The important hydrolysis results from both high precipitation (>1000 mm/year) and high temperatures (>5 °C in winter and >25 °C in summer) that characterize this region. The north-eastern United States as well as the south-western Mississippi River watershed display minimum values of the (S + K)/(I + C) ratio (<1). As expected from local climatic conditions and altitude, these areas seem to be dominated by physical weathering processes. The respective contributions of both physical

and chemical weathering can not be easily deduced in the other areas characterized by intermediate values of the (S + K)/(I + C) ratio.

4.2. Distribution in the northern Gulf of Mexico

The clay mineral distribution maps of the Gulf of Mexico cover various geomorphologic provinces including the Texas slope, the Louisiana shelf and continental slope, the Mississippi River delta, the Alabama shelf, the Florida shelf and the deep-sea plain (Figs. 3, 6 and 7).

Smectite is the dominant clay mineral and represents more than 50% of the clay mineral fraction in surface sediments from the central and western parts of the Northern Gulf of Mexico (Table 1, Fig. 6a). The smectite content is lower on the Alabama and Florida shelves with values below 40%. The illite content ranges between 13% and 40% (Fig. 6b). Overall, the illite abundance decreases from the northern Gulf of Mexico continental shelves toward the deep-sea plain. This decreasing trend is particularly clear from the proximal to the distal parts of the Mississippi River Delta. Nevertheless, an illite-rich area extends from the Louisiana coasts to the deep-sea plain and constitutes an exception in the illite general trend. The distribution of kaolinite displays an overall West–East increasing gradient in the Gulf of Mexico (Fig. 7a). Kaolinite percentages are minimal (<20%) on the central and western parts of the northern Gulf of Mexico. They increase eastward and reach heir maximum (>24%) on the Florida shelf. Chlorite is the least abundant clay mineral in the Gulf of Mexico, ranging from 0% to 20% (Fig. 6b). The chlorite content is minimal in the areas located off the Mississippi River mouth and in the Louisiana shelf (<4%), and maximum in the eastern and southern parts of the Northern Gulf of Mexico.

5. Main clay mineral provinces of the United States of America east of the Rocky Mountains

The United States east of the Rockies can be subdivided in four major mineralogical provinces (Fig. 9) based on the distribution maps of the main clay minerals (Fig. 6 and 7).

(Zone 1) The western part of the Mississippi River watershed including the entire Missouri River system, is defined by high smectite content (>40%; Fig. 6a). The relative abundance of smectite reflects the mineralogy of the Great Plains soils (Allen et al., 1972; Smith, 2003). Early studies (Murray and Leininger, 1956; Pinsak and Murray, 1960) noticed that the Missouri River carries large amounts of smectite of bentonitic origin. Smectite in the north-western Mississippi river watershed could be related to aeolian volcanic deposits in

Canadian prairie soils (Curtin et al., 1998). Part of the smectite may also be authigenic and form in poorly drained soils, from the main Mississippi River valley or from the surrounding periglacial loess (Fig. 5; Pye and Johnson, 1988). Illite is less abundant, ranging from 10% to 30% (Fig. 6b). In this province, an increase of illite in the detrital loads of rivers is observed after winter, when illite, formed through glaciogenic processes, by physical weathering and erosion of parent rocks, is easily removed by rainfall/meltwater and then exported towards the ocean via rivers (Weaver, 1967). The eastern part of this area (Wisconsin), in the Upper Mississippi River catchment basin between Lakes Superior and Michigan is characterized by locally higher kaolinite content (>20%; Fig. 7a). Kaolinite is derived commonly from a saprolite developed during the Mesozoic and early Tertiary on Precambrian basement, particularly in the area of River Falls (site 8 in Fig. 4; Irving, 1876; Cummings and Scrivner, 1980).

(Zone 2) The region composed by the Great Lakes area and the eastern Mississippi River watershed is characterized mainly by illite (>25%; Fig. 6b) and chlorite-rich sediments (>20%; Fig. 7b). These minerals result from primarily physical weathering of lower/ middle Palaeozoic rocks (Appalachian Mountains; Griffin et al., 1968; Potter et al., 1975). This distribution pattern agrees with previous observations in the same area (Pinsak and Murray, 1960; Weaver, 1967; Johnson and Kelley, 1984).

(Zone 3) The south-eastern United States region is typified by the relative abundance of kaolinite, often exceeding 40% (Fig. 7a). The erosion of Neogene marine terraces along the Georgia coast, which is covered by red-yellow podzolic soils, derived from the Appalachian Mountain saprolites (Fig. 5), yields kaolinite-rich sediments (Pirkle,1960). The reflectance data of Balsam and Beeson (2003) support the hypothesis that Georgia is one of the main sources of kaolinite delivered to the Gulf of Mexico. The abundance of kaolinite in this province reflects mostly the composition of polygenetic palaeosoils, although produced at a lower rate today.

(Zone 4) The last province covers the south-western Mississippi River- and the Brazos River watersheds (Fig. 9). River clay loads from this area are mainly composed of illite (>50%; Fig. 6b) and kaolinite (>25%; Fig. 7a). High values of illite in this area are explained by the proximity of Colorado Plateau where frost shattering and other mechanical erosion processes reactivate recurrently in wintertime. The Llano Estacado, which is in the middle of this area (Fig. 5), is uplifted plateau of Late Tertiary riverine illiteand kaolinite-rich sediments (Parry and Reeves, 1968; Glass et al., 1973; USGS Geological map).

6. River inputs into the Gulf of Mexico and oceanographic implications

Three processes largely control the distribution of continentally derived clay minerals in the sediments of the Northern Gulf of Mexico.

6.1. River inputs

According to Pinsak and Murray (1960), the main factor controlling the marine distribution of clay minerals is the nature of the dominant particle sources. Our data show that the Northern Gulf of Mexico can be roughly divided into two separated provinces: a western smectite-rich (>50%) and kaolinite-poor (<20%) province and (2) an eastern smectite-poor and kaolinite-rich province (Figs. 6a and 7a). This pattern clearly suggests that the clay mineral distribution in the Northern Gulf of Mexico is controlled by two main river systems.

The western smectite-rich province covers the main part of the Gulf of Mexico. This peculiar distribution reflects the prevailing detrital supply via the Mississippi River since the latter delivers the sediments to the Gulf of Mexico that are the most enriched in smectite (60–80%). This is consistent with the fact that Mississippi River discharge is 20 times larger than all other rivers combined feeding the Gulf of Mexico (Griffin, 1962; Chamley and Kennett, 1976). As a consequence the fine-grained sedimentation in the Northern Gulf of Mexico is mainly controlled by the detrital inputs from the Mississippi River. Since the Mississippi River detrital supply is depleted in chlorite, the area directly under the influence of the Mississippi River inputs is characterized by chlorite-poor deposits (<4%; Fig. 7b).

The eastern kaolinite-rich province corresponds mainly to the Alabama–Florida shelf and the clay mineral assemblages probably signal a local origin. The Apalachicola River and the Mobile River system, which drain both the kaolinite-rich south-eastern continental province (Zone 3) and the chlorite-rich Appalachian Mountains (Zone 2; Fig. 9), likely account for the kaolinite- and chlorite-rich sediments onto this eastern province (Fig. 7). Parker et al. (1992) noted indeed that the provenance of the clay-mineral suite observed in the Alabama shelf is primarily from the Mobile River system. Moreover, Mazzullo and Peterson, 1989 analysed the late Quaternary silts on the Northern Gulf of Mexico continental shelf and identified two provinces in the Alabama–Florida shelf which correspond to areas directly influenced by the Mobile River and Apalachicola River respectively. The coeval occurrence of high kaolinite and chlorite contents in surface sediments of the eastern province of the Gulf of Mexico is somehow surprising. Indeed, chlorite is early removed under the weathering conditions that prevail in Alabama, Florida and Cuba whereas kaolinite is likely to be more resistant. Such characteristics suggest that chlorite and kaolinite must result from at least two

different source rivers (as reflected by different stages of weathering). Unfortunately we have not found data to support this hypothesis yet. Russell et al. (2000) noticed that chlorite is one alteration mineral in the Encrucijada Formation, of Albian–Cenomanian age, west of Havana. This chlorite may be currently (or have been over the recent past) transported to the Gulf of Mexico and may explain the high chlorite content in sediment off Cuba.

Balsam and Beeson (2003) delineated transport and dispersal paths for kaolinite based on the factor analysis of the first derivatives of the percent reflectance data from 186 core-top and grab samples distributed throughout the Gulf of Mexico. They proposed that kaolinite from the Mobile River system is transported across the shelf and into deeper portions of the Gulf. This is in agreement with our observations and may explain why sediments off Mobile Bay and of the Mississippi River Delta have slightly higher kaolinite content (>12%) than sediments of the Louisiana continental slope (Fig. 7a).

The other northern Gulf-Coast Rivers (e.g. Atchafalaya, Brazos, and Rio Grande rivers) have more local influences. Atchafalaya Bay delivers illite-rich sediments to the Louisiana shelf. These sediments are further transported across the Louisiana slope and into the deep Gulf of Mexico (Fig. 6b). The Rio Grande and/or its tributary (the Pecos River) drain the illite- and kaolinite-rich continental province defined as Zone 4 (Fig. 9) and distribute illite-rich sediments (>26%) over the Texas continental shelf and slope (Fig. 6b). Similarly, the clay mineral fraction of sediments off the Rio Grande and into the deep-sea plain are richer in kaolinite than Louisiana continental slope deposits, in agreement with Balsam and Beeson (2003) results. However, the respective kaolinite concentration maps show some discrepancies. According to Balsam and Beeson results (2003), the maximum kaolinite concentration area is located in the deep-sea plain of the Gulf of Mexico. Kaolinite content is highest in sediments of the Alabama–Florida shelf in our distribution map, in agreement with the previous work of Griffin (1962). Moreover, the comparison of kaolinite distribution map on the continent with marine distribution map suggests a clear relationship between the major continental source of kaolinite and the kaolinite-rich Alabama–Florida shelves (Zone 3; Fig. 9).

6.2. Differential settling

A regular offshore decrease of illite contents off the main Northern Gulf-coast Rivers with an exception for the Atchafalaya Bay appears on the illite distribution map (Fig. 6b). Illite generally has the largest grain size distribution of the clay mineral fraction in the Mississippi River suspended sediment (Johnson and Kelley, 1984; their figure 2) and in loess.

Even when illite floccules are smaller than those of smectite, they are denser (Sakamoto, 1972), and consequently, illite settles more rapidly than the other clay minerals. The observed zoned pattern suggest that differential settling, with larger flocs and grain-size being deposited nearest shore, is one of the processes affecting clay mineral distribution in the Northern Gulf of Mexico. The distributions of various clay mineral peak-area ratios (illite/smectite, illite/kaolinite, illite/chlorite [maps not shown]) over the Northern Gulf of Mexico confirm the apparent decrease in abundance of illite towards the deeper basin. This local differential settling of clay minerals has not been previously reported elsewhere in Gulf of Mexico sediments (Pinsak and Murray, 1960; Griffin, 1962; Scafe and Kunze, 1971; Doyle and Sparks, 1980).

6.3. Oceanic processes

In addition to river inputs and differential settling, oceanic currents may also affect the distribution of clay minerals in the Northern Gulf of Mexico sediments. The surface circulation over the Gulf of Mexico, especially along the coast is modulated seasonally by changes in primary wind patterns. The dominant hydrologic feature of the Gulf of Mexico is the clockwise Loop Current (Fig. 1). The Loop Current enters the Gulf through the Yucatan Channel, intrudes variably northward, sometimes as far as the Mississippi Delta, and exits through the Florida Straits. Anti-cyclonic rings (Loop Current Rings) separate from the Loop Current every 6–17 months and travel westward across the Gulf of Mexico (Ohlmann and Niiler, 2005). General circulation on the Louisiana– Texas shelf is dominated by a cyclonic gyre, resulting in a strong, westward, coastal current associated with a relatively weak eastward current along the shelf break.

These hydrological features most likely influence the marine clay mineral distribution patterns in the Gulf of Mexico (Figs. 6 and 7). The geographical extent of the two distinct clay mineral marine provinces (see Section 6.1) is indeed consistent with an anti-cyclonic transport in the north-eastern Gulf of Mexico driven in part by the Loop Current. For instance, an eastward transportation of Mississippi sediments enriched in smectite is apparent on the western Alabama shelf. Similarly, southward transportation of the Apalachicola River kaolinite-rich sedimentary load clearly occurs over the Florida shelf (Figs. 1, 6a and 7a).

In the western part of the Gulf of Mexico, the chlorite-poor Louisiana continental shelf deposits likely result from a westward transportation of the Mississippi River chlorite-poor sedimentary load due to the dominant oceanic currents that affect this area (Fig. 7b). This transport pattern is not clear in the smectite distribution map due to its lower resolution (i.e.

higher range of values). Likewise, the illite-rich suspended sediments from the Rio Grande River are transported eastward over the Texas continental slope under the influence of the prevailing eastward oceanic currents.

The overall distribution of clay mineral assemblages in the Gulf of Mexico is in agreement with previously proposed sediment dispersal paths in the area excepted for the Florida shelf (Balsam and Beeson, 2003; Ellwood et al., 2006).

7. Conclusions

Clay mineral distribution maps of the North American continent and over the Northern Gulf of Mexico were generated using clay mineral data from erodible/alluvial sediments on land and from marine surface sediments. The United States can be divided into four provinces representing the main clay mineral source areas for sediments exported toward the Gulf of Mexico. The north-western Mississippi River watershed is characterized by high smectite content which reflects both the composition of Cretaceous, Tertiary and Pleistocene bedrock associated with a bentonitic or authigenic origin. The Great Lakes area and the Ohio–Tennessee River catchment areas deliver mainly illite and chlorite, due to the chiefly physical, as opposed to chemical, weathering and erosion of Palaeozoic rocks. The Brazos River and south-western Mississippi River watersheds are dominated by illite and kaolinite released by the erosion of the Colorado Plateau and the Llano Estacado. The south-eastern part of the United States constitutes a major kaolinitic province. This kaolinite is derived from inherited red yellow podzolic soils.

The comparison of clay distribution patterns of marine surface sediments with results obtained on the continent permits the identification of three main factors controlling clay-mineral distribution in the Gulf of Mexico. (1) Relative fluvial contribution is the most important factor controlling the clay mineral composition of marine sediments. The dominant source is the terrigenous supply via the Mississippi River as supported by the high smectite content over the Northern Gulf of Mexico, whereas the Mobile, Apalachicola, Atchafalaya, Rio Grande and Brazos Rivers inputs have more local influences. (2) Differential settling, suggested by the illite gradient from the coast to the deep sea off the main Gulf- Coast Rivers, is identified as a contributing factor controlling clay distribution locally in the Gulf of Mexico. (3) Surface oceanic currents, controlled by either the Loop Current or seasonal winds, influence the distribution of clay minerals. We can identify a southward transport of the Mobile and Apalachicola kaolinite-rich sediments over the Florida shelf; a westward transport of Mississippi-derived, chlorite-poor river sediments due to westward currents

affecting the eastern part of the Louisiana shelf and an eastward transport of Rio Grande suspended sediments over the Texas slope. The distribution of the two clay mineral marine provinces (the western smectite-rich province and the eastern kaolinite-rich province) is indeed coherent with the anti-cyclonic north-eastern Gulf of Mexico circulation influenced by the Loop Current. This overview of the dominant factors controlling recent clay mineral distributions in the Gulf of Mexico within the studied transect lays the groundwork for the use of clay-mineral assemblages for downcore investigations, particularly during the last glacial cycle when clay mineral fluvial inputs have likely been different from those of today due to the presence of the Laurentide Ice Sheet which modified the erosional pattern. The study of the clay mineral assemblages supplied to the Gulf of Mexico during meltwater pulses should, for instance, provide valuable information on their provenance, and therefore contribute to documenting the climatic conditions over the North American continent during the last glacial period (Sionneau et al., in preparation).

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Figure captions

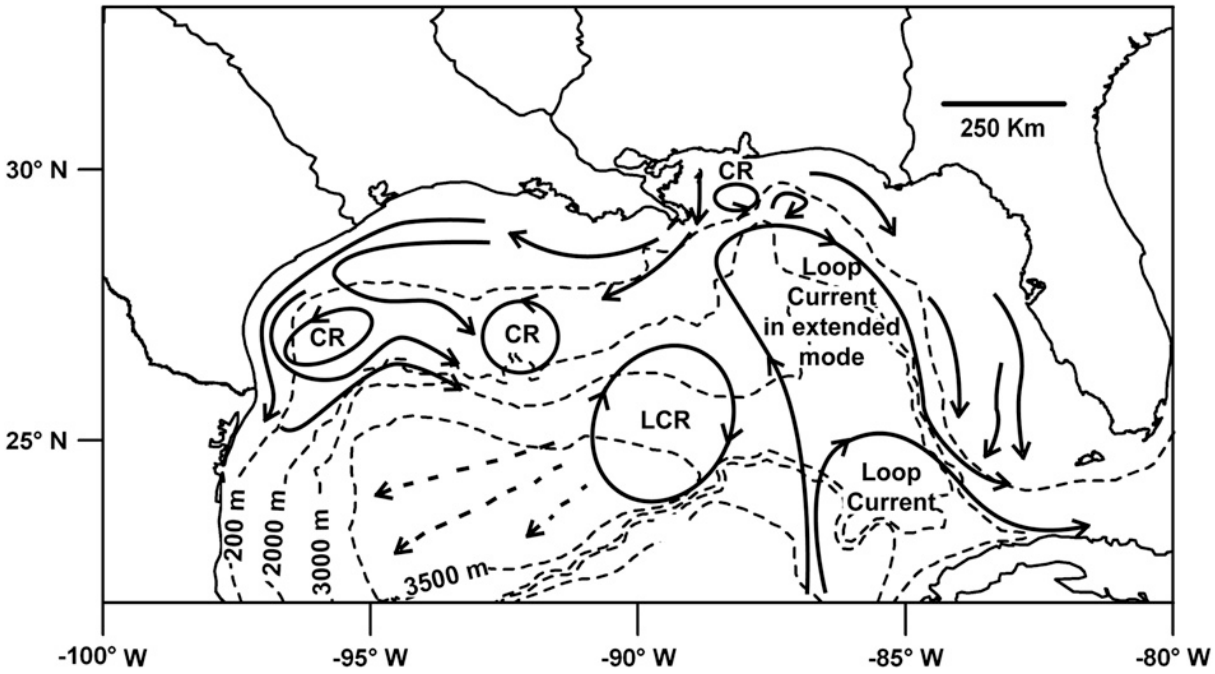
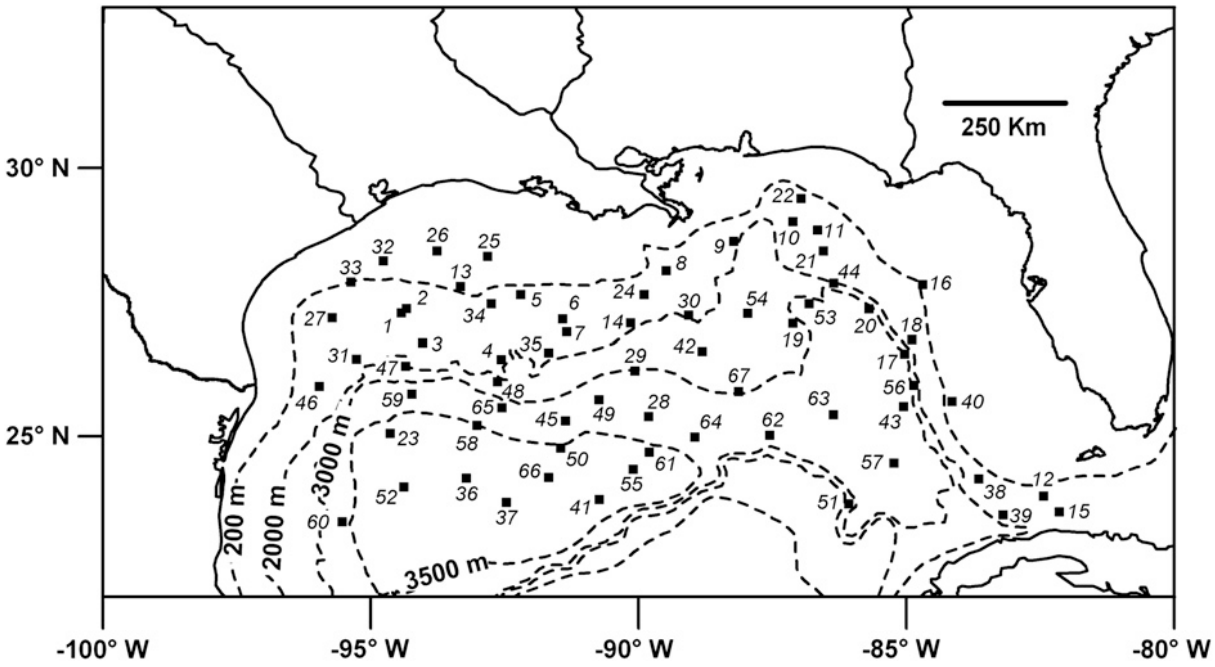


Fig. 1. Mean currents of the Gulf of Mexico (bold black arrows), including two different states of the Loop Current, Loop Current Rings (LCR) and Cyclonic Rings (CR) (adapted from Schmitz, 2003; Fan et al., 2004; Ohlmann and Niiler, 2005; Smith and Jacobs, 2005). Dotted arrows represent propagation direction for LCR (Vukovich and Crissman, 1986).



628

629 Fig. 2. Location of the 67 marine cores (1–67) recovered during RVs Robert Conrad, Vema,

630 Atlantis and Marion Dufresne cruises (RC, VM, AT and MD respectively in Table 1).

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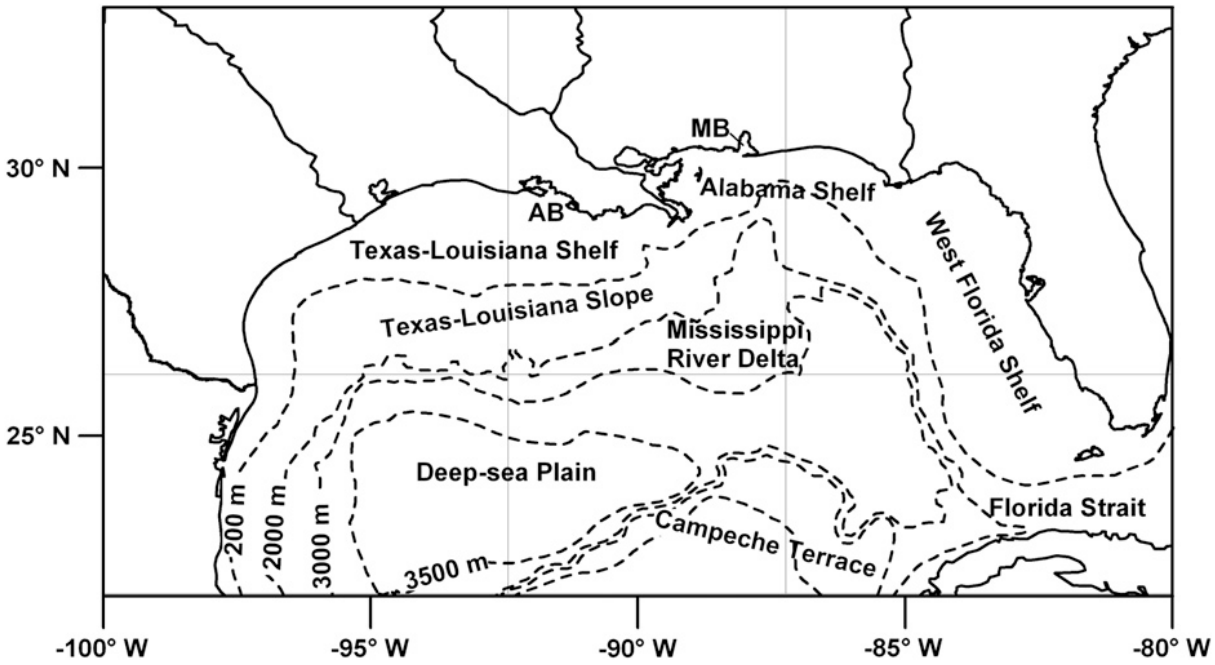
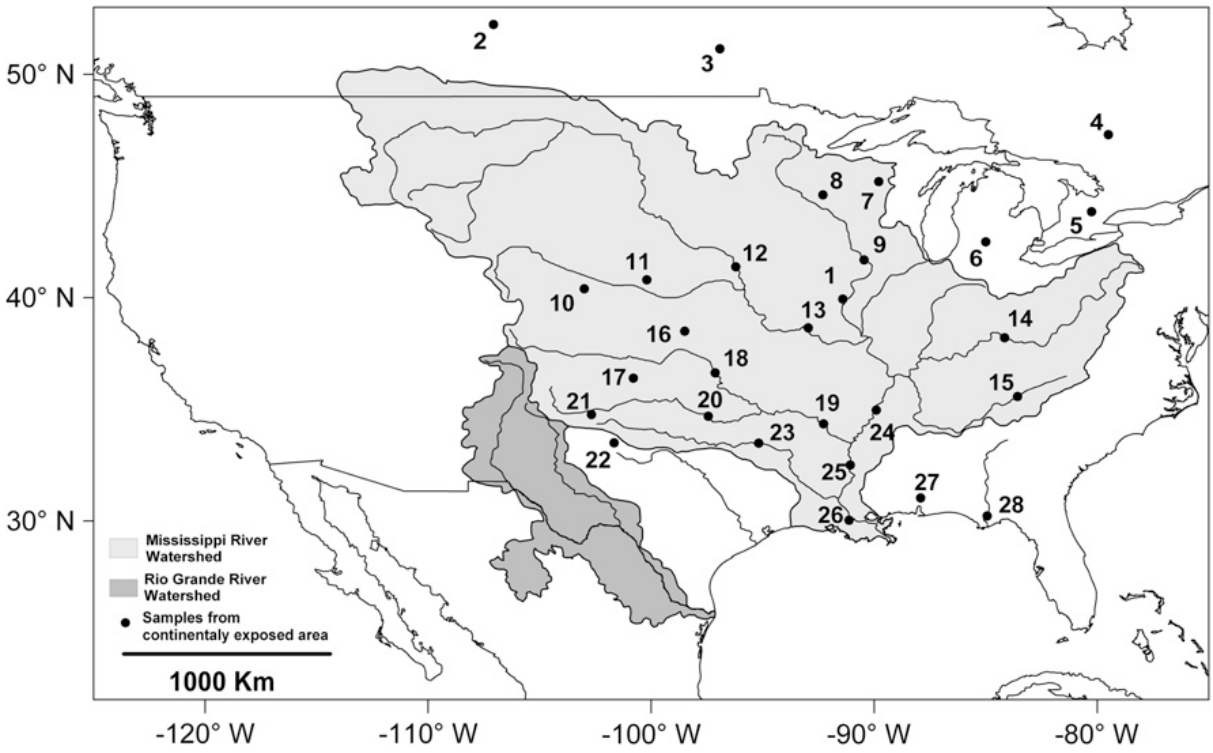


Fig. 3. Geomorphologic provinces of the Gulf of Mexico (AB: Atchafalaya Bay; MB: Mobile Bay).



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636 Fig. 4. Location of the 28 North American (1–28) sites used in the study. The extension of the
637 Mississippi River and Rio Grande River watersheds are respectively represented in light grey
638 and dark grey (Brown et al., 2005; Dahm et al., 2005; Delong, 2005; Galat et al., 2005;
639 Matthews et al., 2005; White et al., 2005).

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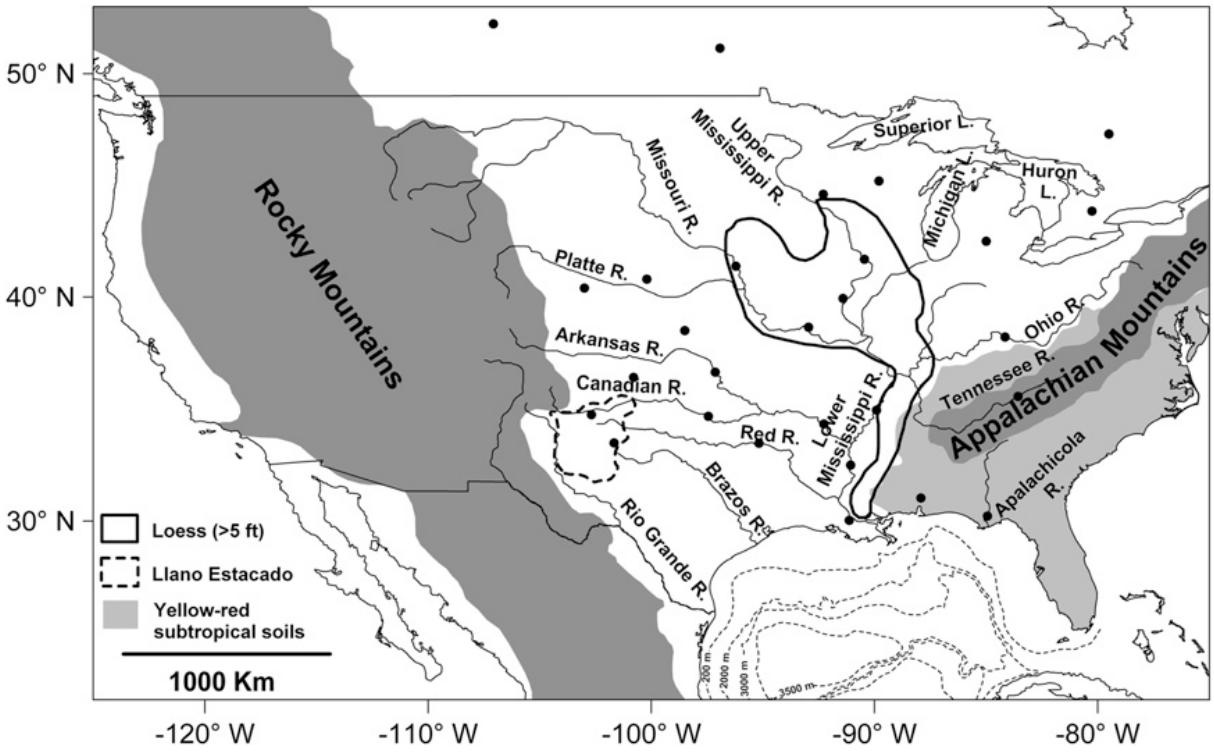
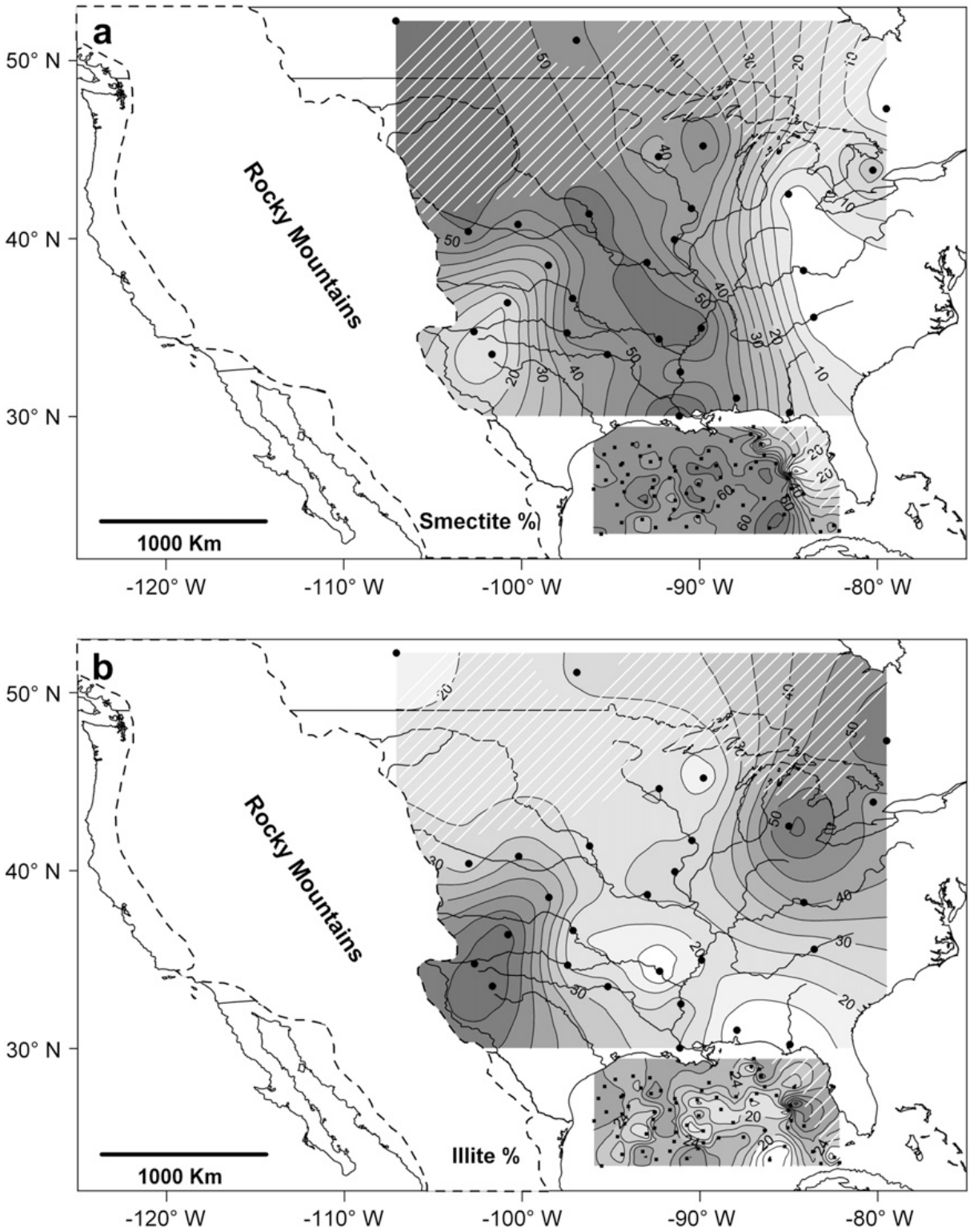


641

642 Fig. 5. Location of rivers and main geological provinces (loess province, yellow-red tropical

643 soils, Llano Estacado, Appalachians and Rocky Mountains).



644

645 Fig. 6. Distribution of smectite (a) and illite (b) over the North American continent east of the

646 Rocky Mountains. White lines indicate areas with larger uncertainties.

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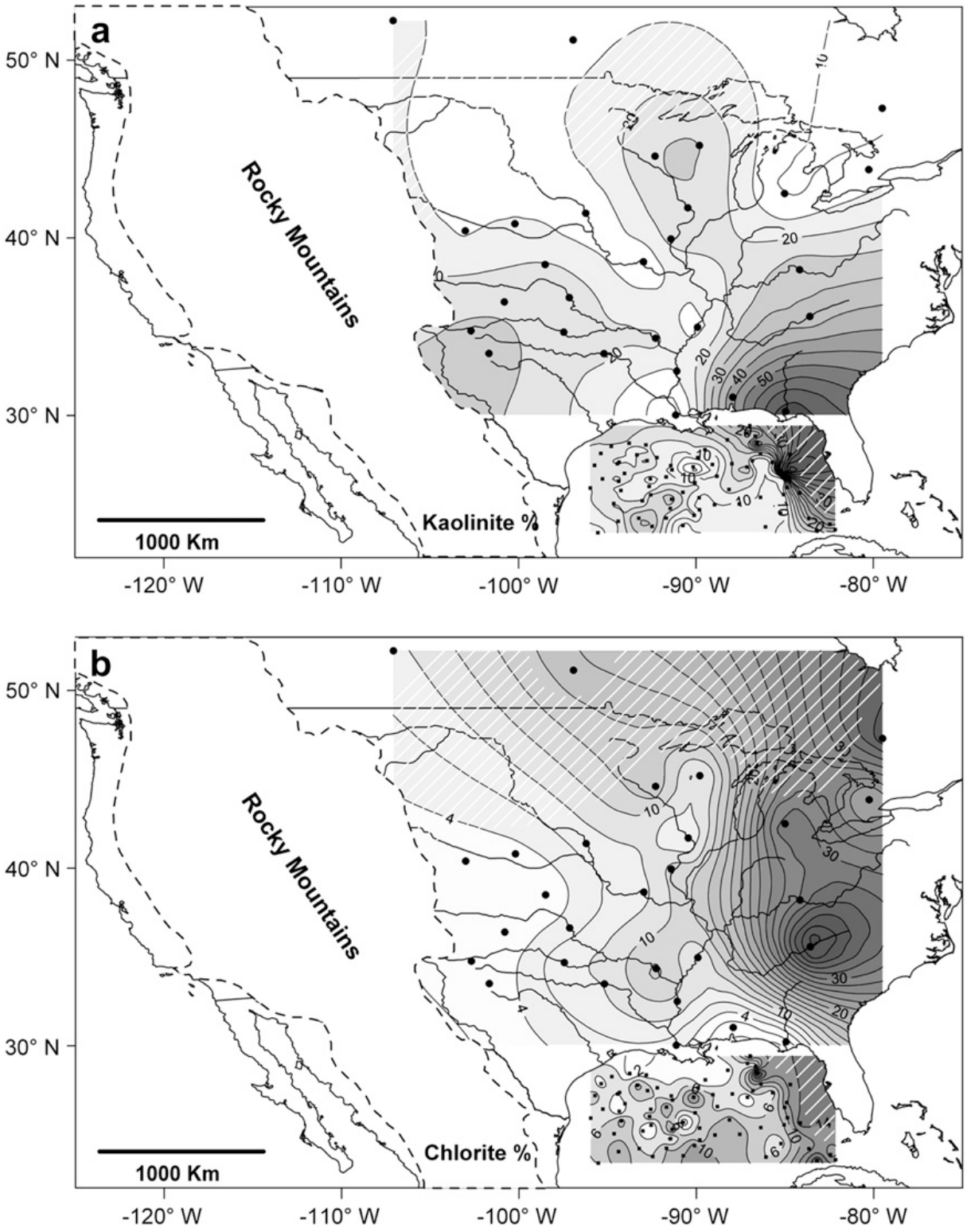


Fig. 7. Distribution of kaolinite (a) and chlorite (b) on the studied area. White lines indicate areas with larger uncertainties.

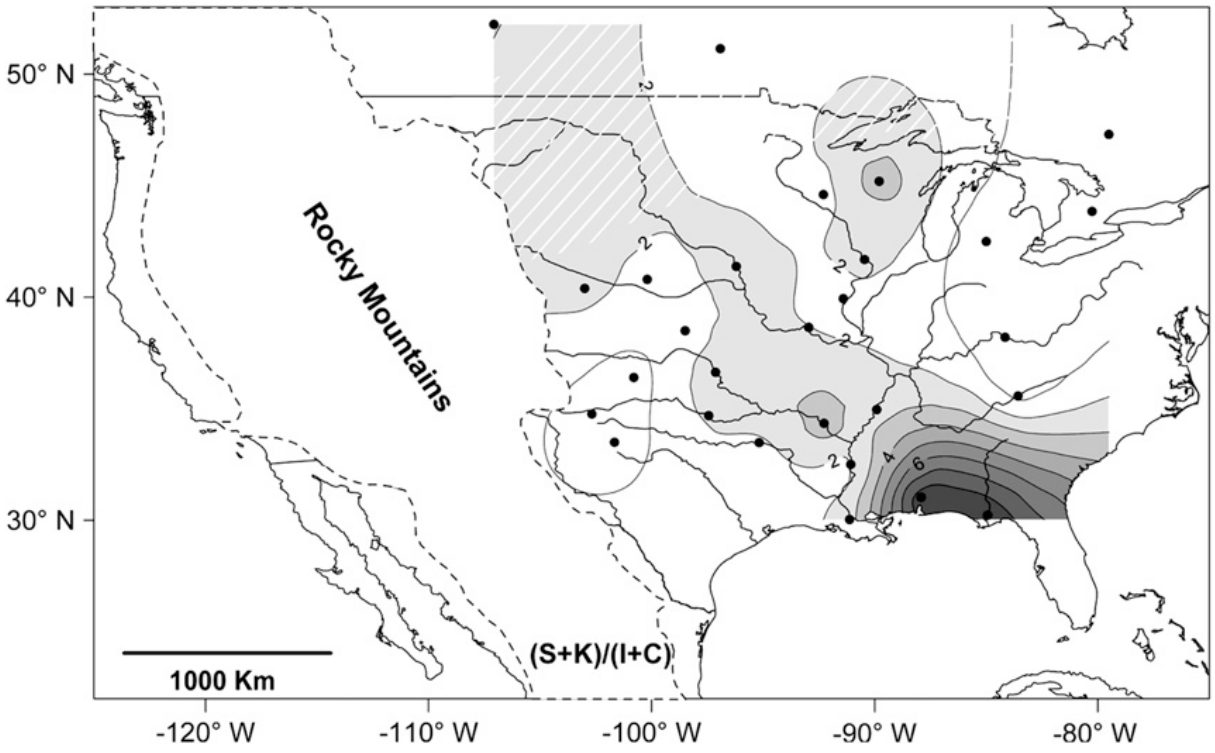
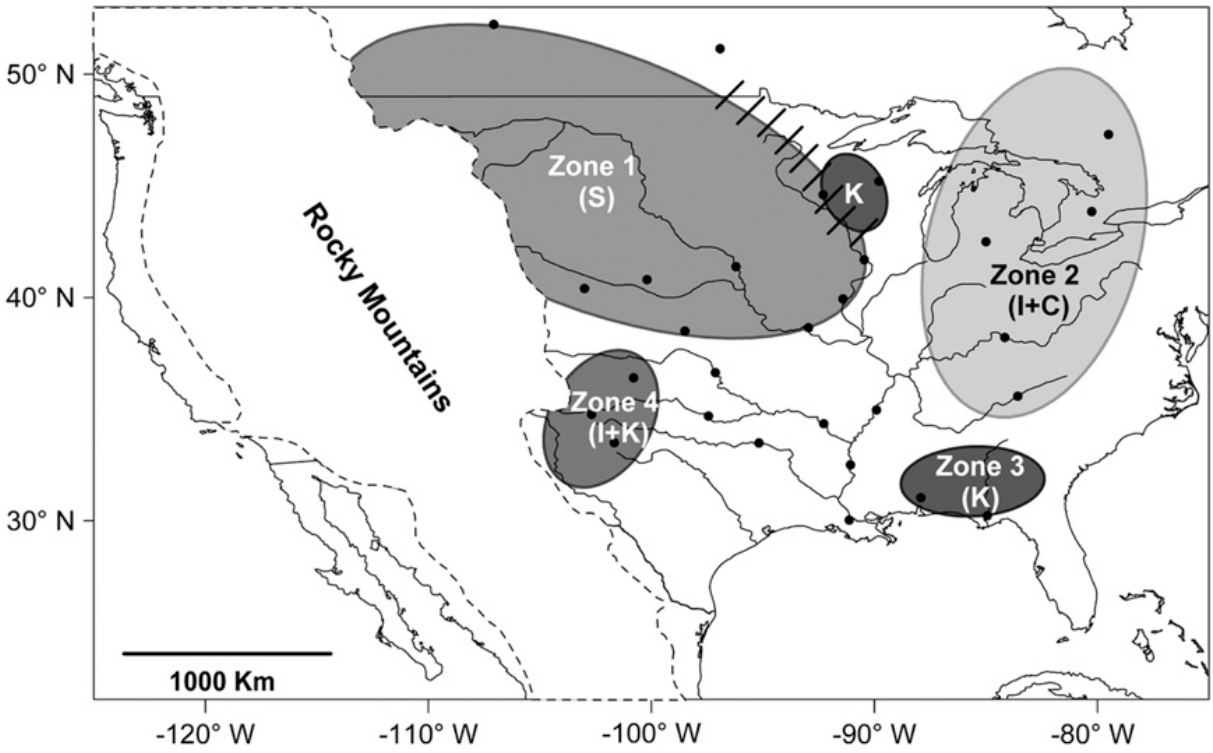


Fig. 8. Distribution of the (smectite + kaolinite)/(illite + chlorite) ratio which indicates the type of weathering (chemical or physical) that affected the erodible sediments. High/low values of this ratio suggest that chemical/physical weathering prevails in the studied region. But the respective contributions of both physical and chemical weathering can not be easily deduced in the areas characterized by intermediate values of the (S + K)/(I + C) ratio. For instance, the values observed in Wisconsin (site 7) reflect the presence of kaolinite inherited from old saprolite layers. In that case intermediate values of the (S + K)/(I + C) ratio, cannot be interpreted as a weathering index. White lines indicate areas with larger uncertainties.



662

663 Fig. 9. Map of the four major clay mineral provinces over the United States. The smectite-rich
664 zone covers a kaolinite-rich area near the Great Lakes (Zone K). Black dashes along the
665 northern border of Zone 1 indicate uncertainty in the position of the boundary between this
666 zone and Zone K.

667

668 Table 1

669 Location, water depth and clay mineral composition of surface sediments from the northern

670 Gulf of Mexico

Site	Core	Latitude	Longitude	Water (m)	Smectite (%)	Illite (%)	Kaolinite (%)	Chlorite (%)
1	MD03-2641	27.29° N	−094.43° W	1427	58	27	11	4
2	MD03-2643	27.37° N	−094.33° W	1379	57	27	12	4
3	MD02-2533/2534	26.74° N	−094.03° W	1350	59	23	14	4
4	MD02-2549	26.43° N	−092.57° W	2049	67	20	9	4
5	MD02-2548	27.64° N	−092.20° W	610	57	28	10	5
6	MD02-2553	27.18° N	−091.42° W	2259	62	23	11	4
7	MD02-2550/2552	26.95° N	−091.35° W	2245	60	25	9	6
8	MD02-2558	28.08° N	−089.49° W	1125	62	25	11	2
9	MD02-2574	28.63° N	−088.22° W	1963	56	25	16	3
10	MD02-2575/2576	29.00° N	−087.12° W	848	60	16	19	5
11	MD02-2577/2578	28.84° N	−086.66° W	476	53	25	18	4
12	MD02-2581	23.88° N	−082.44° W	1063	49	14	26	11
13	AT185-43P	27.78° N	−093.33° W	183	59	23	9	9
14	AT185-58P	27.11° N	−090.15° W	2138	72	15	0	13
15	VM3-7P	23.58° N	−082.15° W	1681	42	25	20	13
16	VM3-9P	27.82° N	−084.70° W	181	34	19	34	13
17	VM3-16P	26.53° N	−085.03° W	3286	62	15	13	10
18	VM3-19P	26.80° N	−084.90° W	730	0	40	48	12
19	VM3-34P	27.10° N	−087.12° W	2889	62	17	19	2
20	VM3-35P	27.37° N	−085.70° W	3221	64	20	11	5
21	VM3-39P	28.45° N	−086.55° W	647	20	30	30	20
22	VM3-42P	29.43° N	−086.97° W	667	32	28	27	13
23	VM3-68P	25.05° N	−094.63° W	3630	55	23	15	7
24	VM3-81P	27.63° N	−089.90° W	1123	57	26	12	5
25	VM3-83P	28.35° N	−092.82° W	149	58	27	12	3
26	VM3-90P	28.45° N	−093.77° W	140	61	24	14	1
27	VM3-93P	27.20° N	−095.72° W	964	52	27	14	7
28	VM3-98P	25.37° N	−089.82° W	3424	67	17	9	7
29	VM3-99P	26.22° N	−090.07° W	3157	54	26	14	6
30	VM3-100P	27.25° N	−089.07° W	2034	67	20	7	6
31	VM3-121P	26.43° N	−095.27° W	1648	54	28	13	5
32	VM3-108P	28.27° N	−094.77° W	148	55	25	14	6
33	VM3-112P	27.87° N	−095.37° W	191	57	27	13	3
34	VM3-114P	27.47° N	−092.75° W	761	44	32	17	7
35	VM3-118P	26.55° N	−091.68° W	2212	51	29	10	10
36	VM3-125P	24.22° N	−093.22° W	3744	43	32	19	6
37	VM3-128P	23.77° N	−092.47° W	3590	53	23	18	6
38	VM3-139P	24.20° N	−083.65° W	1103	41	26	22	11
39	VM3-142P	23.53° N	−083.20° W	1785	42	24	16	18
40	VM17-7P	25.65° N	−084.15° W	154	30	30	25	15
41	VM17-19P	23.82° N	−090.73° W	3706	54	30	6	10
42	VM18-362P	26.58° N	−088.82° W	2577	67	17	11	5
43	RC9-15P	25.56° N	−085.06° W	3341	57	25	10	8
44	RC9-16P	27.85° N	−086.36° W	3111	54	24	13	9
45	RC9-18P	25.29° N	−091.37° W	3374	43	28	18	11
46	RC9-21P	25.93° N	−095.96° W	960	56	27	12	5
47	RC9-22P	26.31° N	−094.35° W	1955	55	28	15	2
48	RC9-23T	26.02° N	−092.63° W	2111	58	25	9	8
49	RC9-24P	25.68° N	−090.74° W	3424	73	14	13	0
50	RC9-25P	24.78° N	−091.45° W	3576	58	24	15	3
51	RC9-31P	23.74° N	−086.08° W	3466	71	13	10	6
52	RC10-267P	24.05° N	−094.38° W	3749	58	20	12	10
53	VM24-19P	27.47° N	−086.82° W	3012	63	19	10	8
54	VM24-20P	27.28° N	−087.97° W	2651	55	26	14	5
55	VM24-21P	24.38° N	−090.10° W	3654	46	30	13	11
56	RC12-5P	25.95° N	−084.87° W	2946	47	27	16	10
57	RC12-4P	24.50° N	−085.23° W	3405	72	15	8	5
58	RC12-8T	25.20° N	−093.02° W	3576	71	15	11	3
59	RC12-9P	25.78° N	−094.23° W	3263	61	21	9	9
60	RC10-10P	23.40° N	−095.53° W	3054	61	24	9	6
61	RC12-13P	24.70° N	−089.80° W	3563	66	19	8	7
62	RC12-14P	25.02° N	−087.55° W	3389	56	26	9	9
63	VM26-131P	25.40° N	−086.37° W	3252	60	23	10	7
64	VM26-132P	24.98° N	−088.95° W	3484	56	25	10	9
65	VM26-138P	25.53° N	−092.55° W	1677	55	28	15	2
66	VM26-141P	24.23° N	−091.68° W	3740	54	25	13	8
67	VM26-142P	25.83° N	−088.13° W	3103	59	19	13	9

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673 Table 2

674 Location, sediment type, number of samples (N) and clay mineral composition of sites from

675 North America

Site	Location	Latitude	Longitude	Type	Smectite (%)	Illite (%)	Kaolinite (%)	Chlorite (%)	Reference
1	Quincy, Illinois	39.94° N	−091.41° W	Loess	39	27	22	11	
2	Saskatchewan	52.23° N	−107.07° W	River	59	17	16	7	
3	Lake Manitoba	51.14° N	−096.92° W	Lake	46	26	13	15	
4	Ottawa River	47.30° N	−079.50° W	River	0	54	8	37	
5	Ontario	43.85° N	−080.26° W	Till	25	43	12	20	
6	St. Francis Lake	42.50° N	−085.00° W	Till	1	58	8	33	
7	Stetsonville	45.20° N	−089.80° W	Till	53	15	26	7	
8	River Falls	44.60° N	−092.30° W	Loess	35	26	25	14	
9	Upper Mississippi River	41.69° N	−090.46° W		45	25	25	5	Potter et al., 1975
10	Kelly section	40.40° N	−103.00° W	Loess	57	26	14	3	
11	Bignell Hill/Eustis sections	40.80° N	−100.20° W	Loess	47	34	16	3	
12	Upper Missouri River	41.38° N	−096.21° W		60	22	11	7	Potter et al., 1975
13	Lower Missouri River	38.65° N	−092.96° W		55	27	11	7	Potter et al., 1975
14	Ohio River	38.21° N	−084.16° W		0	41	29	30	Potter et al., 1975
15	Tennessee River	35.58° N	−083.57° W		0	26	31	43	Potter et al., 1975
16	Barton section	38.50° N	−098.50° W	Loess	35	43	21	2	
17	Texas (Norwood House)	36.40° N	−100.80° W	Dust	14	59	24	2	
18	Arkansas River	36.64° N	−097.14° W		50	21	24	5	Potter et al., 1975
19	Lower Arkansas River	34.35° N	−092.27° W		57	8	22	13	Potter et al., 1975
20	Canadian River	34.69° N	−097.45° W		48	23	21	8	Potter et al., 1975
21	Texas (Hereford)	34.77° N	−102.68° W	Dust	14	56	26	4	
22	Texas (Lubbock)	33.50° N	−101.67° W	Dust	9	60	28	3	
23	Red River	33.49° N	−095.78° W		43	28	21	8	Potter et al., 1975
24	Middle Mississippi River	34.96° N	−089.91° W		60	20	12	8	Potter et al., 1975
25	Mississippi River	32.50° N	−091.07° W		45	30	15	10	Potter et al., 1975
26	Lower Mississippi River	30.04° N	−091.13° W		65 (60–80)	25 (20–30)	5 (+chlorite: 10–20)	5 (minor)	Griffin, 1962
27	Mobile Bay	31.03° N	−087.92° W		45 (40–50)	10 (0–10)	45 (40–50)	0 (trace)	Griffin, 1962
28	Apalachicola River	30.23° N	−084.94° W		20 (0–20)	10 (0–10)	70 (60–80)	0 (trace)	Griffin, 1962

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