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1 Clay mineral distributions in and around the Mississippi River watershed 2 and Northern Gulf of Mexico: sources and transport patterns

3
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12 13 Abstract

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

30 31 1. Introduction

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

34 have been extensively investigated since the 1960s (Biscaye, 1965; Griffin and
35 Goldberg, 1969; Rateev et al., 1969; Gradusov, 1974; Windom, 1976). Studies of clay mineral
36 assemblages provide information on climatic conditions, such as precipitation and runoff
37 patterns over the adjacent continents (Chamley, 1989), as well as on the dynamics of river
38 inputs (Pinsak and Murray, 1960; Scafe and Kunze, 1971; Doyle and Sparks, 1980). Clay
39 minerals can be advected over long distances and then settle far away from their source area,
40 especially if being re-transported in the nepheloid layer (Biscaye and Eittrheim, 1977; Jones,
41 1984). Thus, clay mineral assemblages have also been successfully used to trace oceanic
42 current patterns (Petschick et al., 1996; Fagel et al., 1997; Gingele et al., 2001; Boulay et al.,
43 2005).

44 The Gulf of Mexico has been an important area for global thermohaline circulation
45 and for the global climatic system (Broecker, 1991) especially since the closure of the
46 Isthmus of Panama 4.6 million years ago (Haug and Tiedemann, 1998). Gulf of Mexico
47 surface waters are characterized by high temperature and high salinity that further influence
48 the Gulf Streamwater properties. The hydrological features of the Gulf of Mexico were
49 strongly modified by the inputs of freshwater resulting from melting pulses of the Laurentide
50 Ice Sheet (LIS) during the last glacial cycle and the last deglaciation (Kennett and Shackleton,
51 1975; Leventer et al., 1982; Joyce et al., 1993; Aharon, 2003; Flower et al., 2004). Changes of
52 salinity resulting from these melting pulses are thought to have altered the thermohaline
53 circulation, the global heat exchange and worldwide climatic conditions (Broecker et al.,
54 1990; Bond, 1995; Rahmstorf, 1995; Manabe and Stouffer, 1997). Although the direct
55 palaeoceanographic effects of these freshwater supplies on the Gulf of Mexico have been
56 studied largely using $\delta^{18}\text{O}$ variations, the associated terrigenous inputs (mostly clay minerals;
57 Brown and Kennett, 1998) have been poorly investigated. They may, however, provide
58 information on the varying continental provenance of these freshwater pulses and therefore
59 help close the gap between climate variations over the continent (e.g. the dynamics of the
60 LIS/ proglacial lake overflows and/or increase in precipitation; Broecker et al., 1989; Teller,
61 1995; Clark et al., 2001; Clarke et al., 2003) and the hydrological changes in the Gulf of
62 Mexico. In order to use clay mineral records from the Gulf of Mexico for palaeoclimate
63 reconstructions (Sionneau et al., in preparation), one must have a good knowledge of clay
64 mineral continental sources and of the factors influencing their distribution in the Gulf of
65 Mexico. It seems reasonable to assume that the nature of the main clay mineral provinces in
66 North America has remained unchanged during the late Quaternary. Variations in clay
67 mineral assemblages in the Gulf of Mexico surface sediments are then mainly controlled by

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

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

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

83

84 **2. Samples**

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

91 Over the North American continent, 85 samples were collected from different types of
92 sediment (loess, recent mineral dust deposits, tills, lake and river) at 14 sites (Fig. 4, Table 2).
93 Sediment samples of fluvial origin were taken from the C horizon of alluvial plain soils
94 (about 1.5–2 m depth) from which, except for the carbonate content, the mineralogical
95 composition is not likely to have been disturbed by anthropogenic activity and should reflect
96 the bulk composition of superficial deposits such as loess, alluvium or tills (Gallet et al.,
97 1998). All these continental samples are commonly representative of the regional and/or
98 upstream watershed average mineralogical composition. In addition we have included
99 published clay mineral data from 11 sites on the Mississippi river system (Potter et al., 1975)
100 and from the Mobile Embayment, the Mississippi and the Apalachicola River mouths (Fig. 5,
101 Table 2; Griffin, 1962). The majority of the continental sites (20 of 28) are located in the

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

103

104 **3. Methods**

105 X-ray diffraction analyses (XRD) were performed at the University of Lille and the
106 LDEO (Lamont–Doherty Earth Observatory of Columbia University). In this paper, “clay
107 minerals” refer to the major phyllosilicate minerals within the clay-size fraction (generally
108 <2 mm). They are identified by XRD following the protocol of Holtzapffel (1985). All
109 samples were first decalcified with 0.2 N HCl. The excess acid was removed by repeated
110 washings with distilled water and centrifugations. The clay-size fraction was separated by
111 settling according to Stokes’s law, concentrated by centrifugation, and oriented by wet
112 smearing on glass slides. The analyses were run on a Philips PW 1749 generator (copper
113 anode; 40 kV voltage; 25 mA intensity and 1°2 θ /min speed) at the University of Lille, and on
114 a Philips X’Pert-MPD (q–q goniometer; Cu X-ray tube; Peltier-cooled, high sensitivity,
115 Kevex energy dispersive detector) at the LDEO. A complete X-ray diffraction analysis
116 requires several runs to permit precise mineral identification. Two XRD runs were performed
117 routinely for each sample: (1) air-dried sample (“normal” run) and (2) ethylene-glycol
118 vapour saturation for 12 h (“glycol” run). The analyses were run from 2.49° to 32.49° 2 θ .
119 Each clay mineral is characterized by its basal layer plus interlayer interval (d) as revealed by
120 XRD analysis (Brown and Brindley, 1980). Smectite (“S”) is characterized by a peak at 14 Å
121 on the normal run, which expands to 17 Å after saturation by ethylene glycol. Illite (“I”)
122 presents a basal peak at 10 Å on the two runs. Chlorite (“C”) is characterized by peaks at 14
123 Å (001), 7 Å (002), 4.75 Å (003) and 3.53 Å (004) on the two diffraction spectra. Kaolinite
124 (“K”) is characterized by peaks at 7 Å (001) and 3.58 Å (002) on the normal and glycol runs.
125 To distinguish kaolinite from chlorite, the portion of the spectrum containing the basal peaks
126 of kaolinite (002) and chlorite (004) around 3.55 Å is step-scanned in a high-resolution mode
127 following standard procedures described in detail by Petschick et al. (1996). Semi-
128 quantitative estimation of clay mineral abundances is based on peak areas, weighted by
129 empirically estimated factors (Biscaye, 1965; Biscaye et al., 1997) and summed to 100% (S +
130 I + K + C = 100%). Peak area measurements were realized in the glycol runs using the
131 Macintosh Mac- Diff[®] 4.2.5 software (Petschick, 2000). Results are given in Tables 1 and 2.
132 The error on the reproducibility of measurements is estimated to be $\pm 5\%$ for each clay
133 mineral. The mixed-layered clay minerals identified by Potter et al. (1975) in Ohio River and
134 Tennessee River samples (sites 14 and 15) were not taken into account in the percentages
135 since their exact nature (vermiculite and/or illite–chlorite and/or illite–smectite) is unknown.

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

143

144 **4. Clay mineral distribution**

145 In most continental environments, illite and chlorite are usually inherited from ancient
146 rocks (Palaeozoic, pre-Cambrian) modified by physical or moderate chemical weathering at
147 high latitudes or altitude where Quaternary periglacial climatic conditions prevail. In North
148 America, kaolinite forms through long-term weathering processes during preglacial time
149 (Thiry et al., 1999; Thiry, 2000). It can derive from saprolites as well as from old sedimentary
150 formations and may also form in solfatares in association with smectite (Huertas et al., 1999).
151 Smectite is either derived from volcanic ash layers or authigenic in poorly-drained
152 environments as in alluvial or coastal plains (Kraus, 1999; synthesis in Gerrard et al., 2007).
153 Smectite can also be locally formed in the marine environment through early diagenesis,
154 halmyrolysis (Karpoff et al., 1989), or hydrothermal weathering of volcanic rocks (Chamley,
155 1998).

156

157 *4.1. Clay mineral distributions in and around the Mississippi River watershed*

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

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

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

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

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

189 Since in most continental environments, smectite and kaolinite are generally formed
190 by chemical weathering, while illite and chlorite are mostly inherited from ancient rocks
191 modified by physical or moderate chemical weathering, the clay mineral ratio
192 $(\text{smectite} + \text{kaolinite})/(\text{illite} + \text{chlorite})$ indicates the type of weathering (chemical or physical)
193 that affected the erodible sediments. According to the distribution of this $(S + K)/(I + C)$ ratio
194 (Fig. 8), the North American continent east of the Rockies can be divided in different
195 dominant weathering-regime areas.

196 The south-eastern United States is characterized by maximum values of the ratio (>4),
197 suggesting that chemical weathering prevails in this region. This seems to be in agreement
198 with the presence of yellow-red subtropical soils (Fig. 5). The important hydrolysis results
199 from both high precipitation (>1000 mm/year) and high temperatures (>5 °C in winter and
200 >25 °C in summer) that characterize this region. The north-eastern United States as well as
201 the south-western Mississippi River watershed display minimum values of the $(S + K)/(I + C)$
202 ratio (<1). As expected from local climatic conditions and altitude, these areas seem to be
203 dominated by physical weathering processes. The respective contributions of both physical

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

206

207 *4.2. Distribution in the northern Gulf of Mexico*

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

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

227

228 **5. Main clay mineral provinces of the United States of America east of the Rocky** 229 **Mountains**

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

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

238 Canadian prairie soils (Curtin et al., 1998). Part of the smectite may also be authigenic and
239 form in poorly drained soils, from the main Mississippi River valley or from the surrounding
240 periglacial loess (Fig. 5; Pye and Johnson, 1988). Illite is less abundant, ranging from 10% to
241 30% (Fig. 6b). In this province, an increase of illite in the detrital loads of rivers is observed
242 after winter, when illite, formed through glaciogenic processes, by physical weathering and
243 erosion of parent rocks, is easily removed by rainfall/meltwater and then exported towards the
244 ocean via rivers (Weaver, 1967). The eastern part of this area (Wisconsin), in the Upper
245 Mississippi River catchment basin between Lakes Superior and Michigan is characterized by
246 locally higher kaolinite content (>20%; Fig. 7a). Kaolinite is derived commonly from a
247 saprolite developed during the Mesozoic and early Tertiary on Precambrian basement,
248 particularly in the area of River Falls (site 8 in Fig. 4; Irving, 1876; Cummings and Scrivner,
249 1980).

250 (Zone 2) The region composed by the Great Lakes area and the eastern Mississippi
251 River watershed is characterized mainly by illite (>25%; Fig. 6b) and chlorite-rich sediments
252 (>20%; Fig. 7b). These minerals result from primarily physical weathering of lower/ middle
253 Palaeozoic rocks (Appalachian Mountains; Griffin et al., 1968; Potter et al., 1975). This
254 distribution pattern agrees with previous observations in the same area (Pinsak and Murray,
255 1960; Weaver, 1967; Johnson and Kelley, 1984).

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

264 (Zone 4) The last province covers the south-western Mississippi River- and the Brazos
265 River watersheds (Fig. 9). River clay loads from this area are mainly composed of illite
266 (>50%; Fig. 6b) and kaolinite (>25%; Fig. 7a). High values of illite in this area are explained
267 by the proximity of Colorado Plateau where frost shattering and other mechanical erosion
268 processes reactivate recurrently in wintertime. The Llano Estacado, which is in the middle of
269 this area (Fig. 5), is uplifted plateau of Late Tertiary riverine illite and kaolinite-rich sediments
270 (Parry and Reeves, 1968; Glass et al., 1973; USGS Geological map).

271

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

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

275

276 *6.1. River inputs*

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

283 The western smectite-rich province covers the main part of the Gulf of Mexico. This
284 peculiar distribution reflects the prevailing detrital supply via the Mississippi River since the
285 latter delivers the sediments to the Gulf of Mexico that are the most enriched in smectite (60–
286 80%). This is consistent with the fact that Mississippi River discharge is 20 times larger than
287 all other rivers combined feeding the Gulf of Mexico (Griffin, 1962; Chamley and Kennett,
288 1976). As a consequence the fine-grained sedimentation in the Northern Gulf of Mexico is
289 mainly controlled by the detrital inputs from the Mississippi River. Since the Mississippi
290 River detrital supply is depleted in chlorite, the area directly under the influence of the
291 Mississippi River inputs is characterized by chlorite-poor deposits (<4%; Fig. 7b).

292 The eastern kaolinite-rich province corresponds mainly to the Alabama–Florida shelf
293 and the clay mineral assemblages probably signal a local origin. The Apalachicola River and
294 the Mobile River system, which drain both the kaolinite-rich south-eastern continental
295 province (Zone 3) and the chlorite-rich Appalachian Mountains (Zone 2; Fig. 9), likely
296 account for the kaolinite- and chlorite-rich sediments onto this eastern province (Fig. 7).
297 Parker et al. (1992) noted indeed that the provenance of the clay-mineral suite observed in the
298 Alabama shelf is primarily from the Mobile River system. Moreover, Mazzullo and Peterson,
299 1989 analysed the late Quaternary silts on the Northern Gulf of Mexico continental shelf and
300 identified two provinces in the Alabama–Florida shelf which correspond to areas directly
301 influenced by the Mobile River and Apalachicola River respectively. The coeval occurrence
302 of high kaolinite and chlorite contents in surface sediments of the eastern province of the Gulf
303 of Mexico is somehow surprising. Indeed, chlorite is early removed under the weathering
304 conditions that prevail in Alabama, Florida and Cuba whereas kaolinite is likely to be more
305 resistant. Such characteristics suggest that chlorite and kaolinite must result from at least two

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

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

318 The other northern Gulf-Coast Rivers (e.g. Atchafalaya, Brazos, and Rio Grande
319 rivers) have more local influences. Atchafalaya Bay delivers illite-rich sediments to the
320 Louisiana shelf. These sediments are further transported across the Louisiana slope and into
321 the deep Gulf of Mexico (Fig. 6b). The Rio Grande and/or its tributary (the Pecos River) drain
322 the illite- and kaolinite-rich continental province defined as Zone 4 (Fig. 9) and distribute
323 illite-rich sediments (>26%) over the Texas continental shelf and slope (Fig. 6b). Similarly,
324 the clay mineral fraction of sediments off the Rio Grande and into the deep-sea plain are
325 richer in kaolinite than Louisiana continental slope deposits, in agreement with Balsam and
326 Beeson (2003) results. However, the respective kaolinite concentration maps show some
327 discrepancies. According to Balsam and Beeson results (2003), the maximum kaolinite
328 concentration area is located in the deep-sea plain of the Gulf of Mexico. Kaolinite content is
329 highest in sediments of the Alabama–Florida shelf in our distribution map, in agreement with
330 the previous work of Griffin (1962). Moreover, the comparison of kaolinite distribution map
331 on the continent with marine distribution map suggests a clear relationship between the major
332 continental source of kaolinite and the kaolinite-rich Alabama–Florida shelves (Zone 3; Fig.
333 9).

334

335 *6.2. Differential settling*

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

340 Even when illite floccules are smaller than those of smectite, they are denser (Sakamoto,
341 1972), and consequently, illite settles more rapidly than the other clay minerals. The observed
342 zoned pattern suggest that differential settling, with larger flocs and grain-size being
343 deposited nearest shore, is one of the processes affecting clay mineral distribution in the
344 Northern Gulf of Mexico. The distributions of various clay mineral peak-area ratios
345 (illite/smectite, illite/kaolinite, illite/chlorite [maps not shown]) over the Northern Gulf of
346 Mexico confirm the apparent decrease in abundance of illite towards the deeper basin. This
347 local differential settling of clay minerals has not been previously reported elsewhere in Gulf
348 of Mexico sediments (Pinsak and Murray, 1960; Griffin, 1962; Scafe and Kunze, 1971; Doyle
349 and Sparks, 1980).

350

351 **6.3. Oceanic processes**

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

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

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

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

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

380

381 **7. Conclusions**

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

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

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

421

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430

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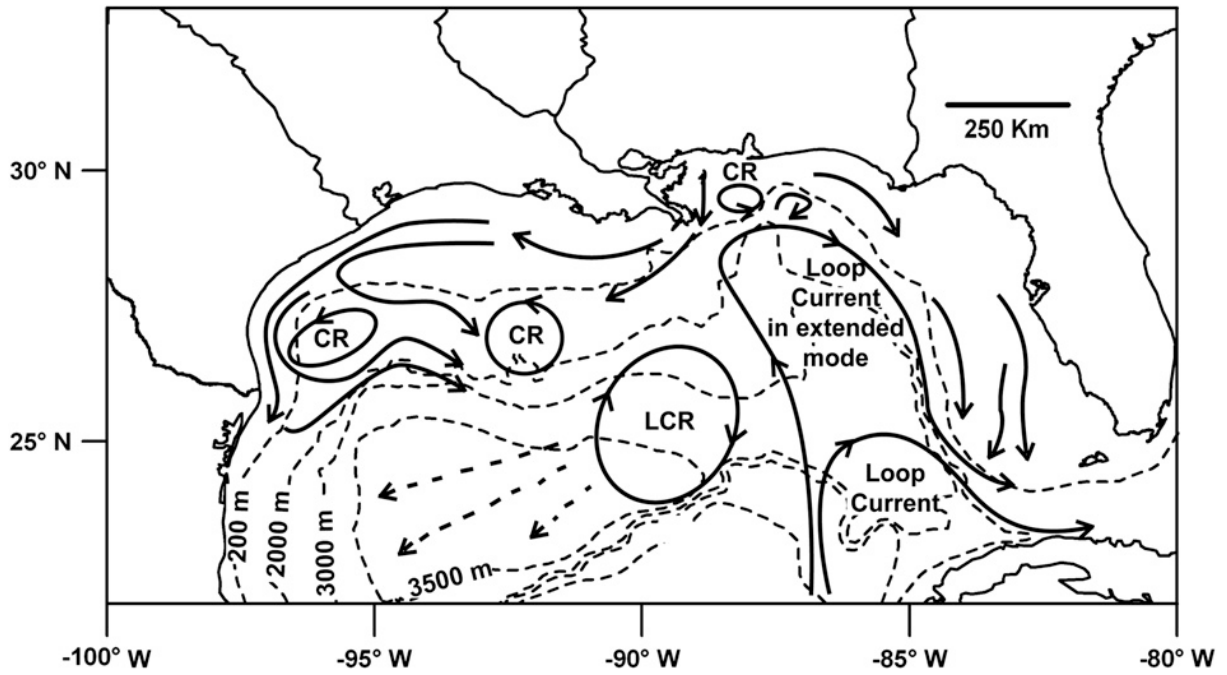
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620 **Figure captions**

621

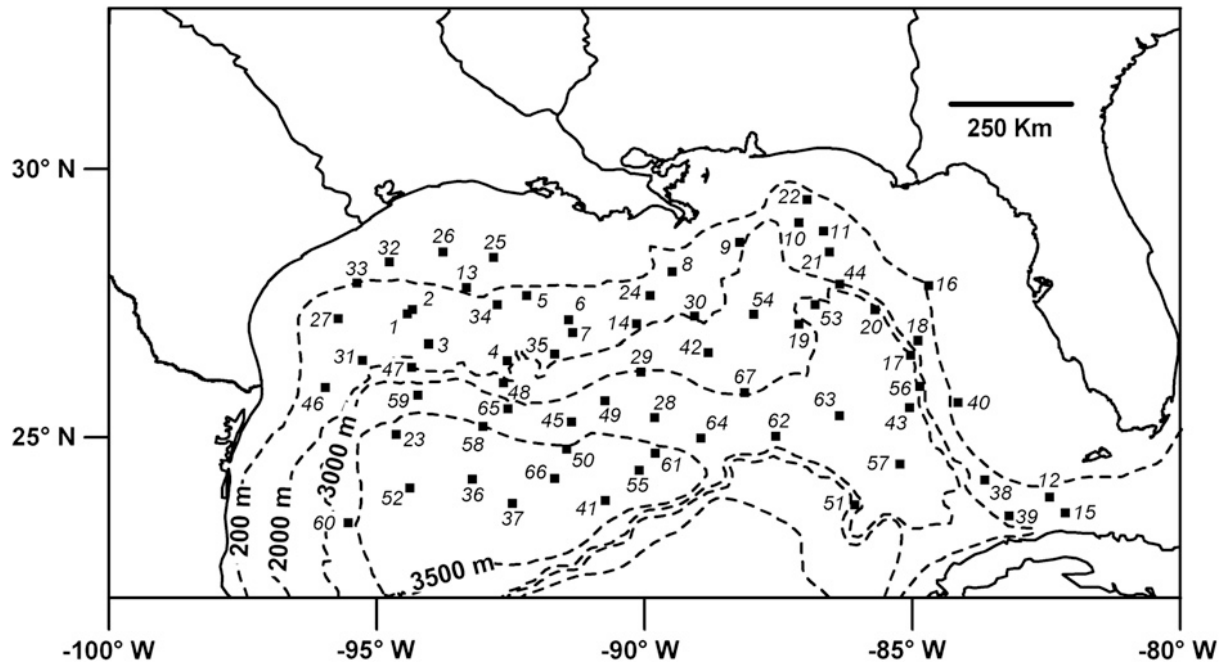


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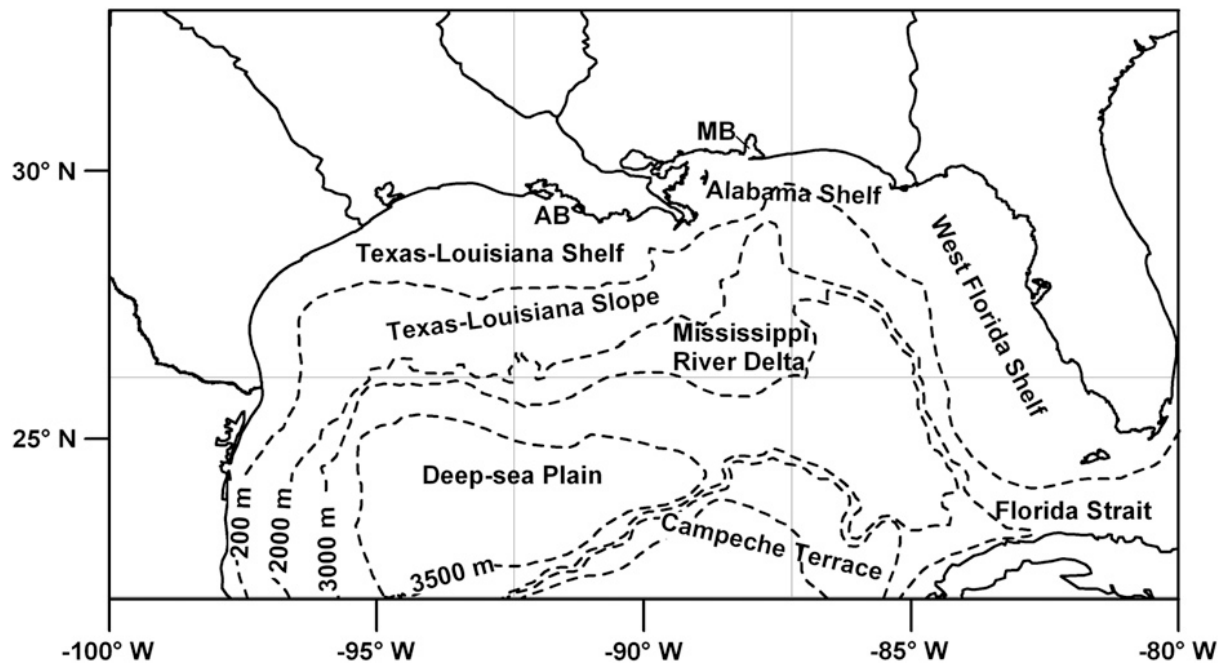
623 Fig. 1. Mean currents of the Gulf of Mexico (bold black arrows), including two different
624 states of the Loop Current, Loop Current Rings (LCR) and Cyclonic Rings (CR) (adapted
625 from Schmitz, 2003; Fan et al., 2004; Ohlmann and Niiler, 2005; Smith and Jacobs, 2005).

626 Dotted arrows represent propagation direction for LCR (Vukovich and Crissman, 1986).

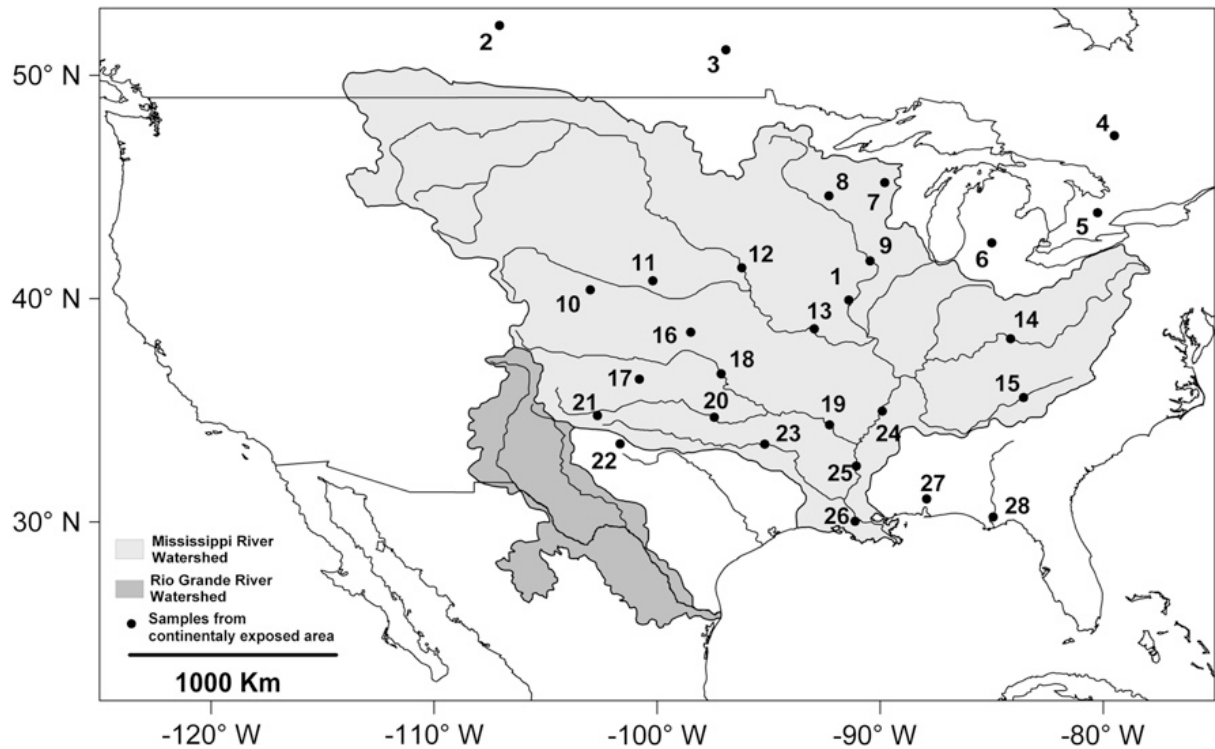
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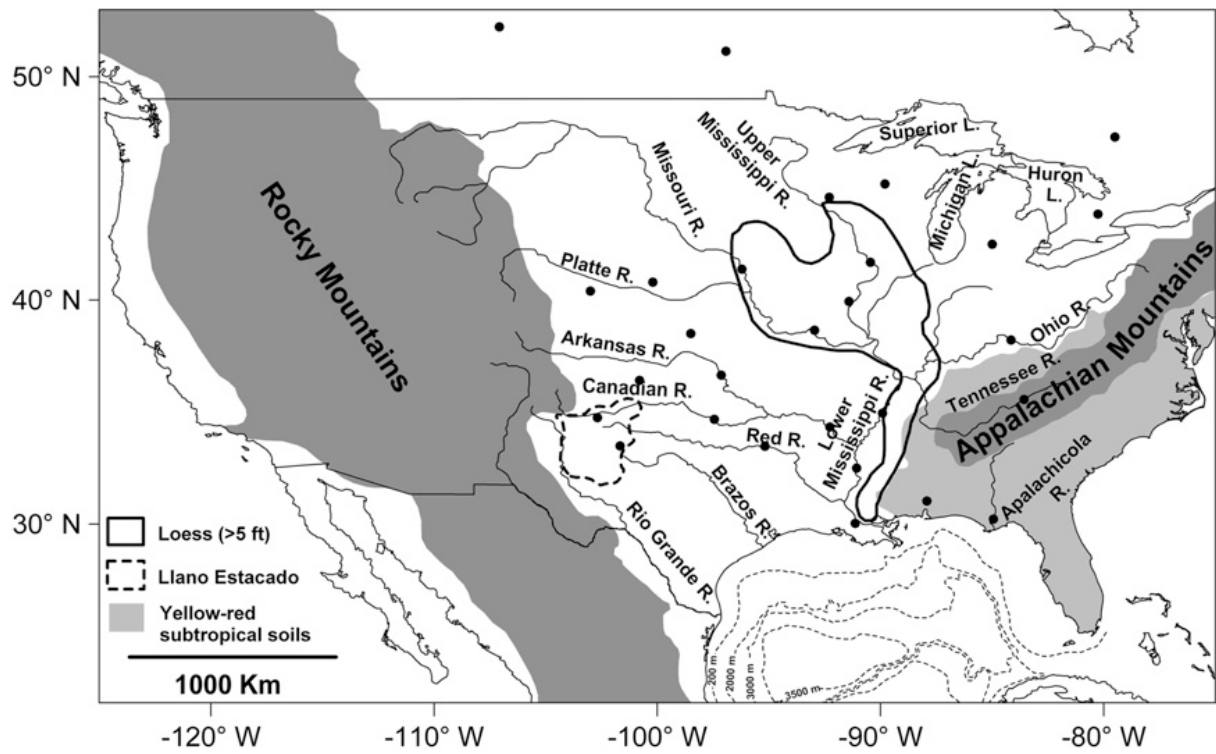
628
 629 Fig. 2. Location of the 67 marine cores (1–67) recovered during RVs Robert Conrad, Vema,
 630 Atlantis and Marion Dufresne cruises (RC, VM, AT and MD respectively in Table 1).
 631



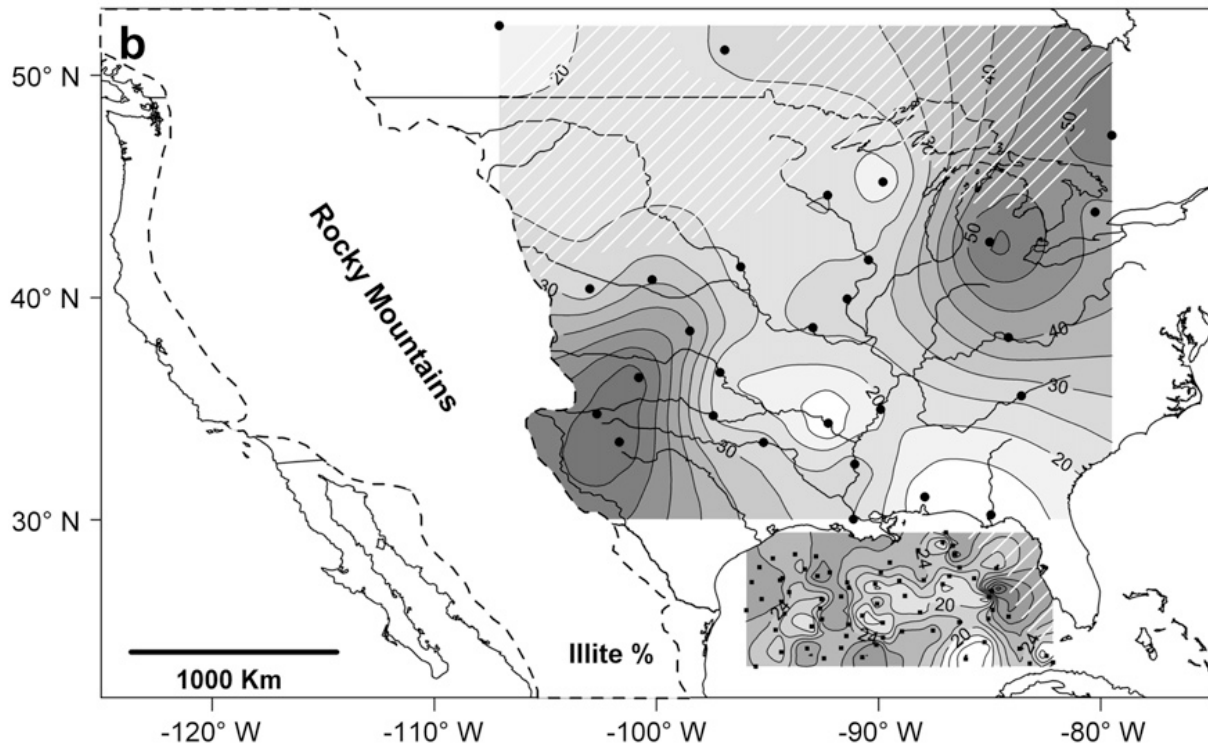
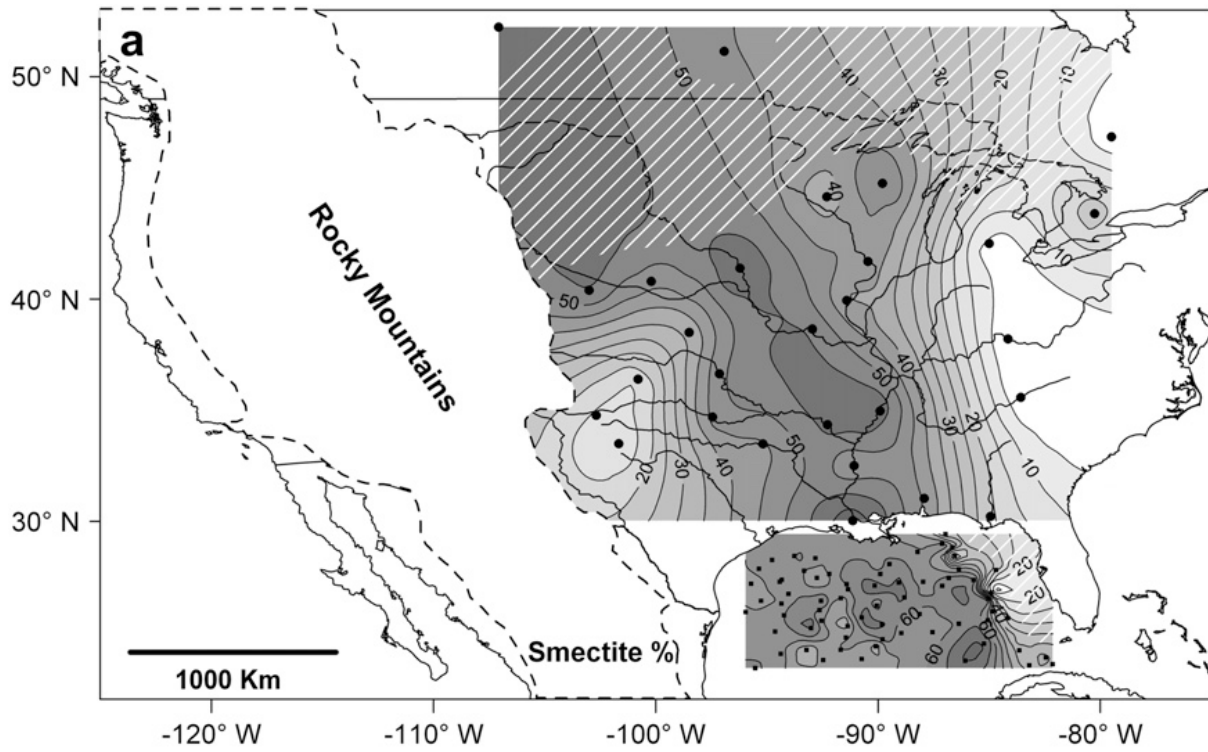
632
 633 Fig. 3. Geomorphologic provinces of the Gulf of Mexico (AB: Atchafalaya Bay; MB: Mobile
 634 Bay).



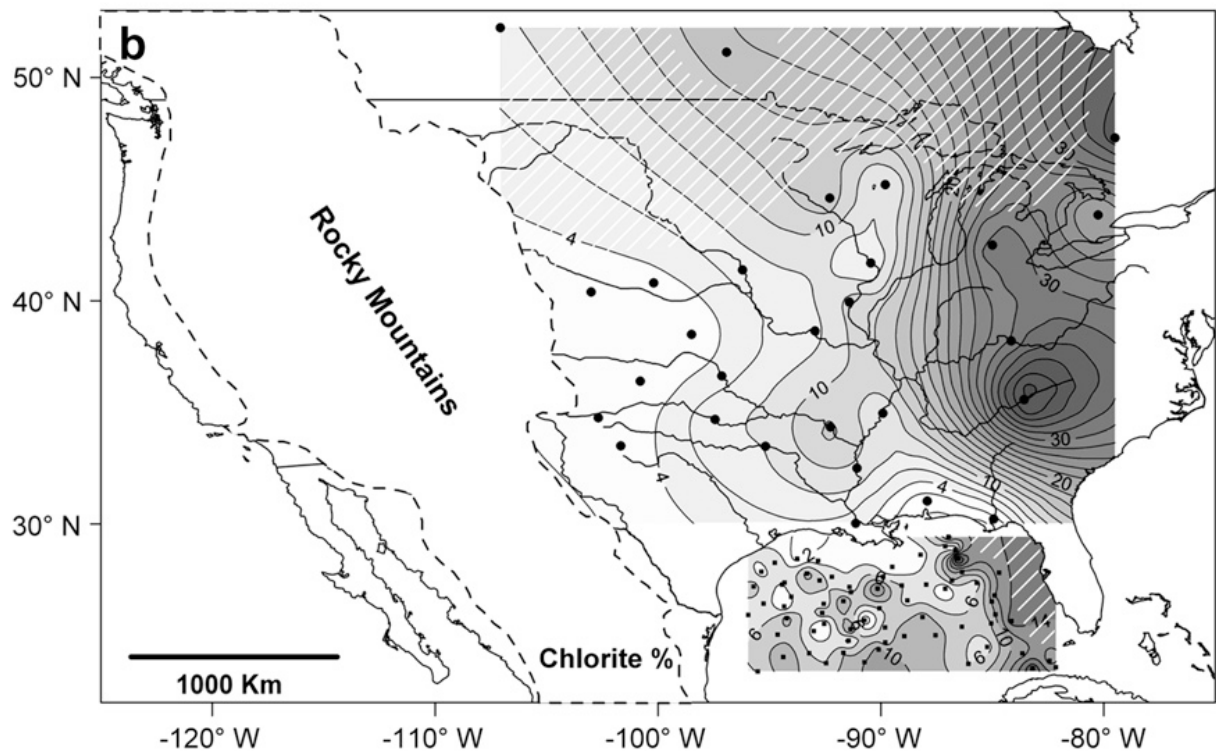
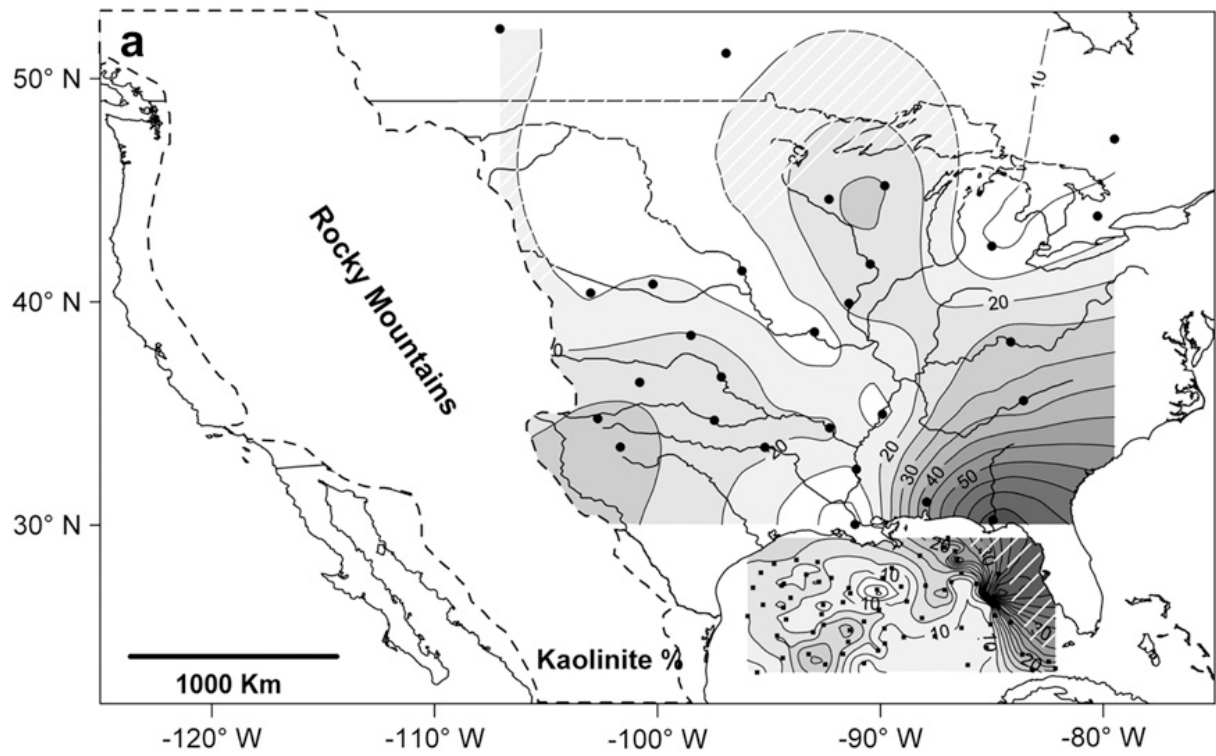
635
 636 Fig. 4. Location of the 28 North American (1–28) sites used in the study. The extension of the
 637 Mississippi River and Rio Grande River watersheds are respectively represented in light grey
 638 and dark grey (Brown et al., 2005; Dahm et al., 2005; Delong, 2005; Galat et al., 2005;
 639 Matthews et al., 2005; White et al., 2005).
 640



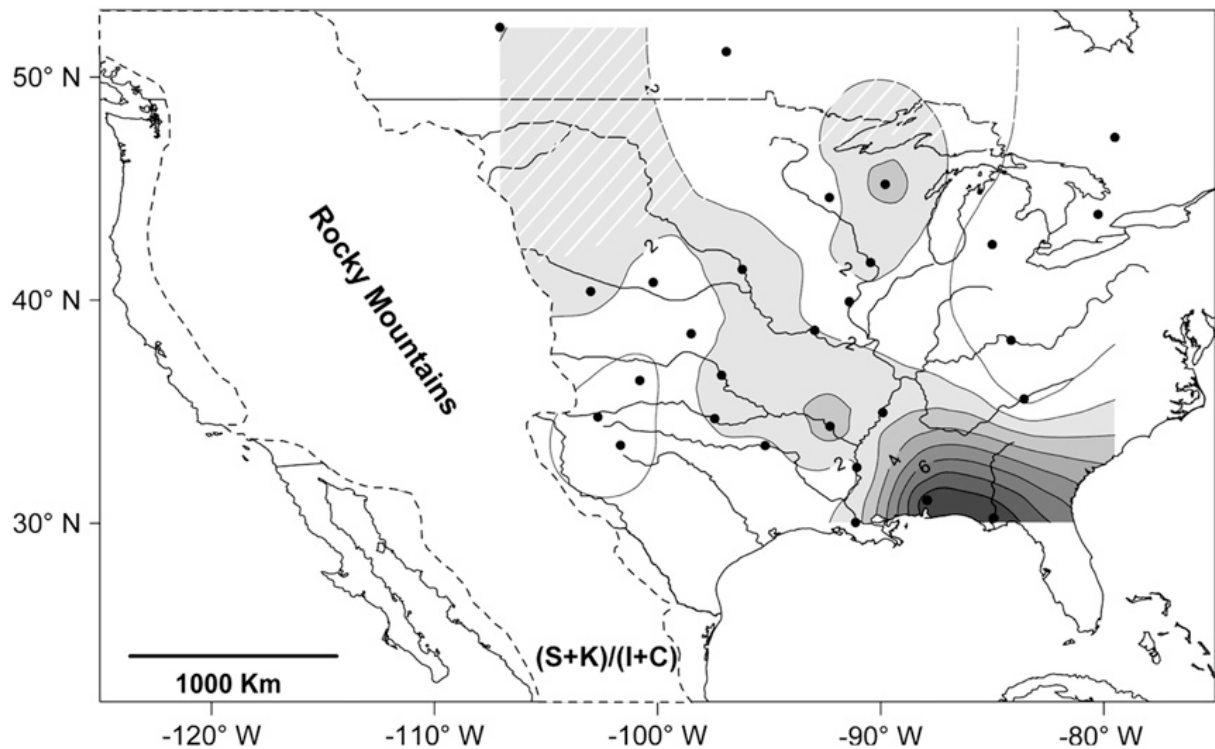
641
 642 Fig. 5. Location of rivers and main geological provinces (loess province, yellow-red tropical
 643 soils, Llano Estacado, Appalachians and Rocky Mountains).



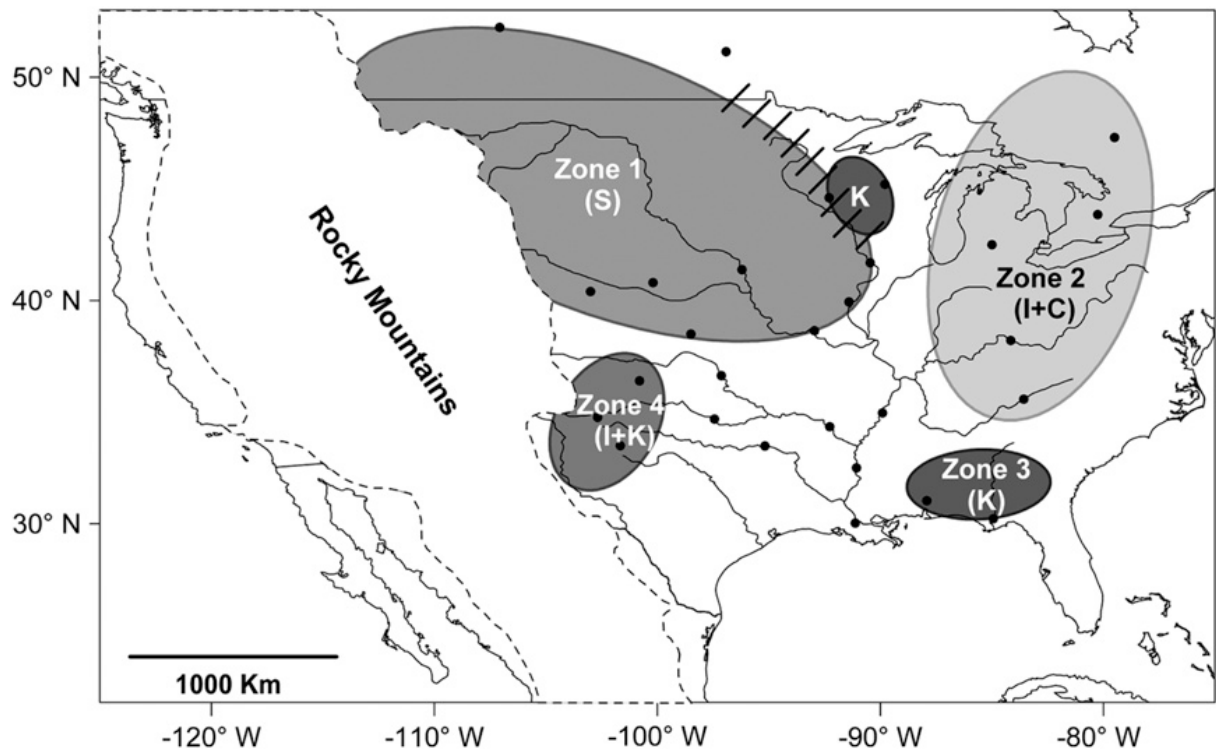
644
 645 Fig. 6. Distribution of smectite (a) and illite (b) over the North American continent east of the
 646 Rocky Mountains. White lines indicate areas with larger uncertainties.
 647



648
 649 Fig. 7. Distribution of kaolinite (a) and chlorite (b) on the studied area. White lines indicate
 650 areas with larger uncertainties.
 651



652
 653 Fig. 8. Distribution of the (smectite + kaolinite)/(illite + chlorite) ratio which indicates the
 654 type of weathering (chemical or physical) that affected the erodible sediments. High/low
 655 values of this ratio suggest that chemical/physical weathering prevails in the studied region.
 656 But the respective contributions of both physical and chemical weathering can not be easily
 657 deduced in the areas characterized by intermediate values of the $(S + K)/(I + C)$ ratio. For
 658 instance, the values observed in Wisconsin (site 7) reflect the presence of kaolinite inherited
 659 from old saprolite layers. In that case intermediate values of the $(S + K)/(I + C)$ ratio, cannot
 660 be interpreted as a weathering index. White lines indicate areas with larger uncertainties.
 661



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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