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# 1 **Controls on detrital sedimentation in the Cariaco Basin during the last** 2 **climatic cycle: insight from clay minerals**

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9  
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11 sedimentation

## 12 13 **Abstract**

14 A high resolution analysis of the clay mineral content of sediments of the Cariaco Basin  
15 spanning the last climatic cycle was performed in order to determine how the detrital  
16 sedimentation in the basin records climatic oscillations at various timescales. At the scale of  
17 glacial interglacial cycles, the clay mineral content records changes in the geographic origin  
18 of the detrital supply, from a dominantly kaolinite- and smectite-rich southern source during  
19 glacial to a more illite-rich northerly source during interglacials. Though possibly influenced  
20 by eustatic variations, these changes mostly reflect the latitudinal fluctuations of the  
21 intertropical convergence zone (ITCZ). The Eemian shows an increased contribution from the  
22 northern source compared to the Holocene. During MIS3, seasonality modulates the effect of  
23 ITCZ position and leads to increased smectite contribution. Fluctuations of the smectite and  
24 kaolinite contents during Dansgaard/Oeschger cycles reflect changes of runoff intensity.  
25 Enhanced supplies in kaolinite during Heinrich-like events and stadials also suggest  
26 remobilizations of sediments deposited on the Unare platform resulting from rapid sea-level  
27 variations. The Younger Dryas is characterized by a significantly different clay mineral  
28 association reflecting a drastic modification of terrigenous supply. An eolian contribution  
29 would explain this peculiar mineralogy, although local contributions cannot be ruled out. A  
30 similar event is noted at the MIS6-5 transition pointing for the occurrence of a Younger Dryas  
31 like episode during Termination II.

32  
33 1. Introduction

34 Tropics are a major source of methane and water vapor to the atmosphere. Through  
35 the emission of these greenhouse gases and of latent heat, tropics play a key role in the  
36 equilibration of the climate system (Ruddiman, 2001; Chiang, 2009). For this reason, tropics  
37 have also been important in the propagation of past climatic variations (Lea et al., 2000;  
38 Hoerling et al., 2001; Ivanochko et al., 2005; Broecker and Putnam, 2013). One of the key  
39 aspects in tropical paleoclimatology is the location of the intertropical convergence zone  
40 (ITCZ). It is well documented that the yearly latitudinal fluctuations of the ITCZ are  
41 superimposed over longer term fluctuations that accompanied larger scale climatic evolutions  
42 from the orbital glacial/interglacial scale, to the pluriannual El Niño Southern oscillations  
43 (e.g., deMenocal et al., 2000b; Wang et al., 2004; Fleitmann et al., 2007; Hessler et al., 2010).  
44 At a given date, location of the ITCZ results from the influence of insolation, landmass  
45 distribution, ice cover and sea surface temperature (SST) distribution, explaining its  
46 sensitivity to global climatic fluctuations (Chiang et al., 2002, 2003; Chiang and Bitz, 2005).  
47 Conversely, the location of the ITCZ has an influence on tropical water budget and can  
48 influence global climate through feedback effects and potential modification of thermohaline  
49 circulation (Romanova et al., 2004; Braconnot et al., 2007; Leduc et al., 2007).

50 Determining past position of the ITCZ using the geological record is difficult. Most  
51 studies are based on proxies that indirectly document changes in rainfall or wind intensities,  
52 allowing to determine whether the study site was located under or beyond the influence of  
53 ITCZ rainbelt or trade winds (deMenocal et al., 2000b; Haug et al., 2001; Wang et al., 2001;  
54 Lachniet et al., 2004, 2009; Jullien et al., 2007; Placzek et al., 2013). The presence (or  
55 absence) of rain in one place mainly documents the position of the summer ITCZ at the study  
56 site. Seasonality and duration of the humid season, which are key parameters for rainfall  
57 distribution and water budget, are however rarely documented. The comparison of records  
58 from different latitudes and in particular from both hemispheres (Wang et al., 2004;  
59 Fleitmann et al., 2007; Lachniet et al., 2009; Arbuszewski et al., 2013) can help trace  
60 latitudinal fluctuations and seasonality, however such an approach needs to compare  
61 extremely well dated records.

62 The Cariaco Basin located off Venezuelan coasts is directly influenced by latitudinal  
63 variations of the ITCZ. During Northern Hemisphere winter, strong trade winds due to the  
64 southern position of the ITCZ lead to arid conditions on land and upwelling of nutrient-rich  
65 water in the basin. From December to April, the sedimentation is therefore dominated by  
66 biogenic opal, carbonate and organic matter (Muller-Karger et al., 2001; Martinez et al.,  
67 2007). During Northern Hemisphere summer, the ITCZ lies above the Cariaco Basin. The

68 decrease of trade-wind intensity leads to significant weakening of the upwelling and  
69 important rainfall leads to a sedimentation dominated by clayey detrital material from June to  
70 October (Muller-Karger et al., 2001; Martinez et al., 2007). Anoxia of the water column below  
71 300m allows the preservation of these seasonal changes as varves: a light colored lamina  
72 deposited during the upwelling season alternates with a dark colored lamina deposited during  
73 the rainy season (Peterson et al., 1991; Hughen et al., 1996a). Due to their strong dependence  
74 on the ITCZ location and finely preserved record, the Cariaco Basin sediments have been  
75 extensively studied during the last decades. In particular, the sedimentary record of latitudinal  
76 fluctuations of the ITCZ on different timescales has been investigated using different  
77 methods: bulk geochemistry (Peterson et al., 2000a; Haug et al., 2001; Peterson and Haug,  
78 2006), sediment color (Hughen et al., 1996b; Deplazes et al., 2013), foraminifer content  
79 (Black et al., 1999), SST paleothermometry based on foraminifers (Lea et al., 2003) and  
80 alkenones (Goni et al., 2006), or palynology (González et al., 2008a, 2008b). Several studies  
81 however also document that sea level variations at different timescales exerted a control on  
82 sedimentation in the Cariaco Basin (Peterson et al., 1991; Clayton et al., 1999; Yarincik et al.,  
83 2000; González and Dupont, 2009), so that deconvolution of the eustatic and climatic signals  
84 may be difficult.

85         The present paper describes the analysis of the clay mineral content of the sediments  
86 from the Cariaco Basin spanning the entire climatic cycle. Because clay minerals mostly  
87 originate from the weathering of emerged lands and are transported to sedimentary basins,  
88 their study allows tracing numerous different processes such as continental weathering  
89 (Chamley, 1989), strength and route of oceanic currents (Fagel et al., 1996, 2001; Bout-  
90 Roumazeilles et al., 1999), rainfall patterns (Montero-Serrano et al., 2009, 2011), or eolian  
91 processes (Jullien et al., 2007) which are key parameters for understanding past variations of  
92 the climate. In the Cariaco Basin sediments, the clay mineral content is used in order to better  
93 constrain temporal changes of the detrital flux in relation to eustatic and climatic fluctuations.

94

## 95 2. Material and methods

### 96 2.1. Studied material

97         The present study is based on sediment from the IMAGES Program core MD03-2625  
98 (10°40.650 N, 64°58.240 W, water depth 847m) recovered during the PICASSO cruise (R/V  
99 Marion Dufresne; Laj and Shipboard Scientific Party, 2004) and Ocean Drilling Program  
100 (ODP) site 1002 (10°42.360 N, 65°10.160 W; water depth, 893 m; Shipboard Scientific Party,  
101 1997). Sediment from holes 1002D and E were used in order to get the most complete record

102 (Hughen et al., 2004). Both coring sites are located on the central saddle of the Cariaco Basin,  
103 however core MD03-2625 is located on its eastern side while ODP site 1002 is located on its  
104 western side (Fig. 1). The presented data correspond to a composite of the two cores: MD03-  
105 2625 for the interval 0e11.66 ka and ODP 1002 for the interval 11.60e132.55 ka. Continuity  
106 of the record was ascertained by the colorimetric correlation of the different cores (see  
107 below).

108

## 109 2.2. Age model

110 The ODP depth scale does not allow a perfect correlation of the different ODP 1002  
111 holes. A new composite depth scale was therefore established by correlating the parameters  
112  $L^*$  (sediment lightness) and the magnetic susceptibility of cores 1002C, 1002D and 1002E  
113 (Table S1; Fig. S1). The age model was then derived from the age models established by  
114 Hughen et al. (2006) and Drenzek (2007), using the new composite depth scale. Rapidly, for  
115 sediment younger than 75 ka (Hughen et al., 2006), the age model is based on the correlation  
116 of core 1002D sediment reflectance with  $^{230}\text{Th}$ -dated stalagmite  $\delta^{18}\text{O}$  records from Hulu  
117 Cave, China (Wang et al., 2001). For sediment older than 75 ka, the age model is based on a  
118 correlation of the oxygen isotopic composition of planktonic foraminifer *Globigerinoides*  
119 *ruber* shells collected from core 1002D (Drenzek, 2007), with the global benthic foraminiferal  
120  $\delta^{18}\text{O}$  stack of Lisiecki and Raymo (2005).

121 For core MD03-2625, the age model was determined by correlating the colorimetric  
122 parameter  $a^*$ , describing sediment color from red to green, acquired on board (Laj and  
123 Shipboard Scientific Party, 2004) with gray-scale data of core PL07-56 PC (Hughen et al.,  
124 1996b), using a more recent age model (Hughen et al., 2000; Table S2; Fig. S2). For the  
125 correlation, core PL07-56 PC was preferred over core ODP 1002, because it is located closer  
126 to core MD03-2625 and is very well dated (Hughen et al., 1996b, 2000). The software used  
127 for the correlation was AnalySeries (Paillard et al., 1996). The obtained ages are calibrated  
128 years before present (yr cal. B.P.). The sedimentation rate thus obtained for the two cores  
129 varies between 0.11 and 1.2 mm/yr with an average value of 0.37 mm/yr for the studied  
130 interval.

131

## 132 2.3. Methods

133 The sediment was systematically sampled with a 10 cm step for the entire climatic  
134 cycle, representing a total set of 480 samples (54 for MD03-2625 and 426 for ODP 1002).  
135 This corresponds to an average sampling interval of 212 yr for MD03-2625 and 286 yr for

136 ODP 1002. Clay mineral associations were studied using X-ray diffraction following the  
137 protocol of Bout-Roumazelles et al. (1999). “Clay minerals” here refer to the major  
138 phyllosilicate minerals within the clay-size fraction (<2-mm particles). All samples were first  
139 decalcified with 0.2 M HCl. Deflocculation of clays was achieved by successive washing with  
140 distilled water. The clay-size fraction was separated by settling according to Stokes’s law,  
141 concentrated by centrifugation, and oriented by wet smearing on glass slides. X-ray diagrams  
142 were obtained using a Philips PW 1749 diffractometer with CuK $\alpha$  radiation and a Ni filter. A  
143 tube voltage of 40 kV and a tube current of 25 mA were used. Three X-ray diagrams were  
144 performed: air-dried sample (normal run), ethylene glycol vapor saturation for 12 h (glycol  
145 run) and heating at 490 °C during 2 h (heating run). The goniometer scanned from 2.49° to  
146 32.49° 2 theta for normal and glycol run and from 2.49° to 14.5° 2 theta for heating run. Each  
147 clay mineral is characterized by its basal layer plus interlayer interval (d) as revealed by XRD  
148 analysis (Brown and Brindley, 1980). Expandable mineral are characterized by a peak at 14Å,  
149 which expands to 15-17Å after ethylene glycol saturation and retracts down to 10Å after  
150 heating. Distinction between smectite (S) and illite-smectite mixed-layer minerals (IS) was  
151 based on the position of the main peak after glycol saturation (17Å and 15Å respectively) and  
152 on the presence of “superstructure” at 29Å for the air-dried sample, expanding at 30Å after  
153 glycol saturation, which characterizes the presence of regular illite-smectite mixed-layer  
154 minerals. Illite (I) is characterized by peaks at 10Å, 5Å and 3.33Å, which remain unchanged  
155 on the three tests. Chlorite (C) is characterized by peaks at 14Å, 7Å, 4.75Å and 3.53Å on the  
156 three runs. Kaolinite (K) is characterized by peaks at 7Å and 3.58Å on the normal and glycol  
157 runs. Both peaks disappear or are strongly reduced after heating. To distinguish kaolinite from  
158 chlorite, the portion of the spectrum containing the basal peaks of kaolinite and chlorite  
159 around 3.55Å is step-scanned in a high-resolution mode following standard procedures  
160 described in detail by Petschick et al. (1996). Semi-quantitative estimation of clay-mineral  
161 abundance is based on peak areas [17Å (smectite) + 15Å (illite-smectite mixed-layers) + 10Å  
162 (illite) + 7.2Å (kaolinite and chlorite)] as measured on the ethylene glycol saturated test using  
163 MacDiff<sup>®</sup> 4.2.5 software (Petschick, 2002) summed to 100%. The error on the reproducibility  
164 of measurements is estimated to be  $\pm 5\%$  for each clay mineral. The illite crystallinity and the  
165 Esquevin index were measured on the XRD diagrams. The illite crystallinity (in °2 theta)  
166 corresponds to the full width at half maximum (FWHM) measured on the illite peak at 10Å  
167 (Chamley, 1989). The illite crystallinity, or Kübler index, is inversely proportional to the  
168 metamorphism degree: a high index indicates very low-to-low metamorphism whereas low  
169 values reflect a high metamorphism. A low signal-to-noise ratio did not allow to correctly

170 determine the Kübler index for core MD03-2625. The Esquevin index or chemical weathering  
171 index (Esquevin, 1969) corresponds to the ratio between the illite intensity measured at 5Å  
172 and at 10Å and is used to evaluate the chemical composition of illite (Al-rich versus Fe/Mg-  
173 rich).

174 Grain-size analysis was carried out on the carbonate-free fraction of the sediment  
175 using a Malvern Mastersizer 2000 laser diffractometer, which has a detection range of  
176 0.02e2000 mm. Deflocculation of the samples was done by successive washing with distilled  
177 water after decarbonation of the sediments with 0.2 M HCl. Sample quantity was adjusted  
178 in order to obtain a laser beam obscuration between 12 and 15%. We used a refractive index  
179 (RI) value of 1.544 (quartz) and an absorption value of 0.05. The distribution parameter  
180 reported here is the mean grain size (mm). Relative uncertainty on this value (2s) is 4e6%  
181 (Sperazza et al., 2004).

182

### 183 3. Results

184 On average, the clay minerals assemblage comprises, in decreasing abundance  
185 kaolinite (K, 33%), smectite (S, 25%), illite (I, 21%), chlorite (C, 14%) and mixed layers  
186 illite-smectite (IS, 7%). The clay mineral assemblages display rather important variations at  
187 orbital timescale. Smectite and illite contents are strongly anticorrelated, but the  
188 anticorrelation is less marked between smectite and (I + IS) (Table S3). The Last Glacial  
189 period (MIS 2e4) is characterized by the lowest illite contents (<20%) and high smectite  
190 contentsw28% (Fig. 2; Table 1). The highest smectite content (30%) is observed during  
191 MIS3. Conversely MIS 5e shows high illite (27%) and the lowest smectite contents (17%)  
192 (Fig. 2; Table 1). MIS5a-tod are slightly enriched in kaolinite (>35%) when compared with  
193 the whole record average values. Relatively low smectite contents (w20%) associated to  
194 higher-than-average mixed layer (>10%) characterize MIS1 (Fig. 2). They are particularly  
195 abundant (17%) at the beginning of MIS1: Younger Dryas in hole ODP 1002, end of Younger  
196 Dryas to Preboreal in core MD03-2625 (Fig. 2; Table 1). Kaolinite content shows marked  
197 variation during terminations: high values at the end of MIS6 and MIS2 are followed by low  
198 values at the MIS6-5 transition and Younger Dryas (Fig. 2). High kaolinite contents also  
199 characterize the end of MIS5b, 5a and 4 (Fig. 2). Chlorite only shows minor variations with a  
200 peculiar enrichment during the Bølling/Allerød (B/A) period (Fig. 2). Compared to the global  
201 variations of benthic  $\delta^{18}\text{O}$  (Lisiecki and Raymo, 2005) and to  $\delta^{18}\text{O}$  values of *G. bulloides*  
202 within the sediment (Drenzek, 2007), the smectite and illite content shows a marked  
203 positive/negative correlation with  $\delta^{18}\text{O}$ , whereas chlorite and kaolinite do not show any clear

204 correlation with  $\delta^{18}\text{O}$  (Fig. 2, Table S4).

205 When clay minerals variations between 20 and 70 ka are observed into more detail, the  
206 smectite content tends to be higher during the long ones (GIS 8, 10 and 14e13), and lower  
207 during low latitude equivalents of Heinrich events (Fig. 3), whereas the kaolinite content  
208 generally shows high values at the beginning of Heinrich-like events (Fig. 3). Smectite and  
209 kaolinite are negatively correlated on this restricted portion (Fig. 3; Table S3).

210 For most samples, grain size distribution is mono-modal. The mean grain size is close  
211 to 10  $\mu\text{m}$  and silts (grains  $<63 \mu\text{m}$ ) represent the dominant fraction ( $>90\%$ ) of the  
212 decarbonated sediment. The mean grain size only shows minor variations and is not  
213 significantly correlated with benthic or planktonic  $\delta^{18}\text{O}$  values (Fig. 4, Table S4). Maximum  
214 values are observed during MIS3 and MIS1. For MIS1, the high values can be related to the  
215 presence of diatoms in the sediment. Minimum values of the mean grain size, reflecting an  
216 increased contribution of clay-sized material are observed during MIS4 and MIS2. Drops in  
217 the mean grain size of the sediment also are often coeval with Heinrich-like and cold events  
218 of MIS3 and MIS5 (Fig. 4).

219

## 220 4. Discussion

### 221 4.1. Evolution of clay minerals at the glacial-interglacial timescale

222 A recent study of the distribution of clay minerals on the shelf surrounding the Cariaco  
223 Basin proposed the use of  $I/(C + K)$  and  $I/S$  ratios to distinguish detrital sources (Bout-  
224 Roumazeilles et al., 2013). In the  $I/S$  vs  $I/(C + K)$  diagram, our new data mainly plot in a  
225 mixing triangle between the Unare, Tuy, and Neveri rivers (Fig. 5). The clay minerals  
226 composition of most intervals can thus be modeled by a simple mixing between these sources  
227 (Table 2). Interglacials (Holocene and MIS5) show a higher contribution from the Tuy and  
228 Neveri rivers (Table 2). This interpretation is supported by the variations of Kübler and  
229 Esquevin indexes (Fig. 4). The Tuy and Neveri Rivers transport illite deriving from highly to  
230 slightly metamorphosed rocks and are hence characterized by a higher crystallinity than the  
231 illite delivered by the Unare River, deriving from Cenozoic and Mesozoic sedimentary  
232 formations (Bout- Roumazeilles et al., 2013). Our data indicate that illite minerals are  
233 generally well crystallized and less altered during interglacials than during glacials (Fig. 4),  
234 supporting an increased contribution from the Tuy and Neveri rivers during interglacials. By  
235 contrast, Last Glacial samples (MIS2 to 4) plot close to the Unare River endmember, though  
236 MIS3 data shows a slightly more dispersed pattern (Fig. 5). To summarize, the variations of  
237 the clay mineral assemblage at the glacial-interglacial timescale primarily result from a



238 change in the location of the detrital sources: during glacials the sediment almost exclusively  
239 originate from a southern source (Unare River) while an additional contribution from northern  
240 sources (Tuy and Neveri rivers) occurs during interglacials.

241 Clayton et al. (1999) interpreted the anticorrelation between smectite and illite, and  
242 their link with the global benthic  $\delta^{18}\text{O}$ , as reflecting eustatically-driven changes in the  
243 respective contribution of smectite-rich local rivers vs the allochthonous supply from the  
244 illite-rich Orinoco-Amazon system. However Bout-Roumazeilles et al. (2013) evidenced that  
245 sediments from the Orinoco delta are rather enriched in smectite, which led to revise this  
246 interpretation. From the distribution of clay minerals on the shelf surrounding the Cariaco  
247 Basin, and consistent with geochemical data (Martinez et al., 2010), Bout-Roumazeilles et al.  
248 (2013) proposed that the mineralogical variations observed by Clayton et al. (1999) reflect a  
249 change in the respective contribution of local rivers only.

250 Even considering local supplies only, the observed provenance changes could still be  
251 of eustatic origin. High sea-level stand during interglacials may promote efficient offshore  
252 mixing of the various detrital sources whereas during glacials, the mouth of the Unare river is  
253 very close to the basin edge favoring specific supply from the Unare river to the deep basin.  
254 Changes in river erosion/deposition in relation to sea-level variations may also be involved.  
255 Rivers are highly erosive during sea-level falls, leading to high sediment delivery to the basin  
256 (e.g., Schumm, 1993). While metamorphic rocks in the Northern coastal range of Venezuela  
257 do not constitute a highly erodible substrate, the Mesozoic to Tertiary sedimentary rocks  
258 present in the Unare drainage basin represent softer material. Important sediment delivery  
259 from the Unare River compared to the Tuy River can therefore be expected during glacial sea-  
260 level falls. Conversely, during and shortly after sea level rise, sedimentation in the previously  
261 incised valleys leads to sediment starving in the basin (Schumm, 1993). The sediment flux  
262 from the Unare River could therefore be reduced during interglacial highstands, allowing the  
263 sediments derived from the northern detrital sources to be less diluted. The anticorrelation  
264 between the terrigenous contribution (Yarincik et al., 2000) and the illite-to-smectite ratio  
265 (Fig. S3) used as a tracer of the southern versus northern contributions gives some support to  
266 these mechanisms. However, the highest contribution of smectite (>30%) is observed during  
267 MIS3, whereas the lowest sea-level is observed during MIS2 (Fig. 2). Similarly, the  
268 calculated Unare River contribution is almost constant for MIS5a to d despite sea level  
269 fluctuations up to 25 m during this interval (Fig. 2). This indicates that provenance changes  
270 are not satisfactorily explained by sea-level fluctuations alone.

271 Clay mineral variations may also be directly climatically-driven. The Cariaco Basin

272 lies in the belt of variation of the ITCZ, so that the average position of the northern  
273 Hemisphere summer ITCZ and associated rainfalls influences the detrital flux. Presently, a  
274 seasonal modulation of the terrigenous contribution to the basin occurs, with the highest  
275 detrital discharge during the rainy season (Martinez et al., 2007). In addition, high rainfalls  
276 characterize the North of Venezuela, in particular the drainage basins of the Tuy and Neveri  
277 Rivers (Fig. 6A; Goldbrunner, 1984), which are affected by high runoff. Previous studies  
278 evidenced the influence of the ITCZ on precipitations patterns at orbital timescale (Peterson et  
279 al., 2000a; Haug et al., 2001; Chiang et al., 2003; Broccoli et al., 2006). During interglacials,  
280 the ITCZ has an average northward position and the associated precipitation belt lies over  
281 northern Venezuela (Fig. 6A). This is consistent with the observed enhanced contribution  
282 from the northern detrital sources during the Holocene and MIS5e (Fig. 5, Table 2).  
283 Conversely a southward move of the average position of the ITCZ during glacials favors  
284 rainfalls over the Venezuelan central plains while rainfalls over Northern Venezuela are  
285 reduced (Fig. 6C). As a consequence, a marked increase in the contribution from the Unare  
286 River (contributing up to 100% of the clay mineral association) is observed during the Last  
287 Glacial (Fig. 5, Table 2). This configuration could however also favor the southern part of the  
288 Neveri River drainage basin which is dominated by the same sedimentary rocks as the Unare  
289 River drainage basin. A contribution of the Neveri River with a similar clay association as the  
290 Unare River is therefore possible (Fig. 6C). The better correlation of clay mineral contents  
291 with planktonic  $\delta^{18}\text{O}$  variations (reflecting SST variations) than with the global benthic  $\delta^{18}\text{O}$   
292 variations (reflecting ice-volume and sea-level; Table S4) also gives support to a dominant  
293 climatic influence, as the highest SST is associated to a northward position of the ITCZ  
294 (Chiang et al., 2002; Chiang and Bitz, 2005).

295

#### 296 4.2. Interglacials

297         Though according to Clayton et al. (1999) interglacials clay-mineral contents are  
298 generally similar, our dataset show marked differences between the Holocene and Eemian  
299 (Fig. 2, Table 1). The Eemian sediments are depleted in smectite (17%), and enriched in  
300 kaolinite (35%) and illite (27%). By contrast, the low smectite content (22%) in Holocene  
301 samples is counterbalanced by a higher contribution of IS (11%). As two distinct sediment  
302 cores are used to record the two time intervals MD03-2625 for the Holocene and ODP 1002  
303 for the Eemian-these differences might be related to the different core locations within the  
304 basin. Core MD03-2625 (NE flank of the central saddle, Fig. 1) should be more influenced by  
305 the eastern detrital sources: the Neveri and Manzanares rivers, and potentially the Araya-

306 Margarita region. Consistently, a high contribution of mixed layer IS is observed in Holocene  
307 samples (Fig. 2, Table 1), a clay mineral which is very specific of the eastern detrital sources  
308 (Bout-Roumazeilles et al., 2013). The very high IS content in Holocene sediments points to a  
309 marked contribution from the Manzanares River, however, the low Esquevin index (Fig. 4)  
310 rather points to a contribution from the Araya region (Bout- Roumazeilles et al., 2013). This  
311 latter contribution is also suggested by the distribution of data in the I/S vs I/(C + K) plot (Fig.  
312 5). From the low clay-mineral content of the sediments located in the northeastern platform,  
313 Bout-Roumazeilles et al. (2013) concluded that these sources likely were of minor influence  
314 for the sedimentation in the deep Cariaco Basin. Flooding events and seismic activity along  
315 the El Pilár fault have been proved responsible for depositional events in the eastern Cariaco  
316 sub-basin (Thunell et al., 1999; Lorenzoni et al., 2012). During these events, clay-rich  
317 sediment (Elmore, 2005) remobilized from the eastern side of the Cariaco Basin and/or from  
318 the Gulf of Cariaco is delivered to the deep basin by turbidity currents flowing along the  
319 Manzanares River Canyon (Lorenzoni et al., 2012). The abundant microturbidites in the  
320 sediments of the eastern sub-basin, including on the eastern flank of the central saddle  
321 (Hughen et al., 1996a), confirms the frequent occurrence of small turbiditic events and  
322 suggests that the contribution of this eastern source has been underestimated by Bout-  
323 Roumazeilles et al. (2013). We can thus assume that the increased contribution of IS during  
324 the Holocene partly results from the eastward location of core MD03-2625 compared with the  
325 ODP 1002. Nevertheless, the I/S and I/(C + K) ratios both point out a significant contribution  
326 from the Neveri River (20%) during the Eemian compared to the Holocene (ca 0%, Table 1,  
327 Fig. 5 middle). Such modification of provenance could not result from respective locations of  
328 the two studied cores, because core ODP 1002 (W side of the central saddle, Fig. 1) is located  
329 farther from the Neveri River than core MD03-2625. Moreover, the higher I/S ratio of the  
330 Eemian (1.94) compared to the Holocene (1.04) indicates a severe decrease of the Unare  
331 River contribution during the Eemian. As a whole, these data suggest a higher contribution of  
332 northern Venezuela sources during the Eemian compared with the Holocene (Fig. 6D).

333 The Eemian interglacial was substantially different from the Holocene, with a  
334 generally warmer climate and higher sea level (Kukla et al., 2002; Rohling et al., 2008; Otto-  
335 Bliesner et al., 2013). This difference results from slightly different orbital parameters, which  
336 led to higher mean insolation during the Eemian than during the Holocene. Several  
337 characteristics of the sediments of the Cariaco Basin such as the total organic carbon content,  
338 detrital content,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  of the organic matter, present contrasting values between the  
339 Holocene and Eemian (Haug et al., 1998; Yarincik et al., 2000; Peterson et al., 2000b;

340 Drenzek, 2007). Alkenone data in the Cariaco Basin indicate that Eemian SST was up to 2 °C  
341 warmer than during the Holocene (Hebert and Schuffert, 2000). Models indicate that the  
342 higher mean summer insolation at 10°N during the Eemian compared with the Holocene  
343 resulted in a more northerly position of the ITCZ, enhanced seasonality and more humid  
344 conditions over Northern South America (Nikolova et al., 2013). Our clay mineral data  
345 suggesting enhanced rainfall on the Coastal Range while the Unare River drainage basin  
346 experienced reduced precipitations, give strong support to a northward displacement of the  
347 mean ITCZ location during the Eemian (Fig. 6D).

348 The illite content rapidly decreases at the transition between MIS5e and MIS5d (Fig.  
349 2). The calculated contribution from the northern detrital sources also decreases and the Unare  
350 contribution during MIS5d is comparable to Holocene value (Fig. 5, Table 2). This suggests a  
351 southward move of summer ITCZ at the end of MIS5e to a position close to the Holocene  
352 position (Fig. 6A), though alkenone data still document high SST in the basin during MIS5d  
353 (Hebert and Schuffert, 2000), similarly to other low latitude paleoclimatic records (Kukla et  
354 al., 2002). In that frame, the rapid decrease in illite synchronous with the North Atlantic C26  
355 cold event (Chapman and Shackleton, 1999; Lehman et al., 2002) (Fig. 4), suggests a strong  
356 forcing of high latitude climatic variations on the modulation of ITCZ shifts (Chiang et al.,  
357 2003).

358

#### 359 4.3. 20-70 ka

360 At orbital timescale, the basic glacial-interglacial variations of the terrigenous supply  
361 to the Cariaco Basin, linked to contrasted mean latitudinal position of the ITCZ and/or to sea  
362 level variations (x4.1) do not satisfactorily explain the peculiar clay composition observed  
363 during MIS3. Indeed, while kaolinite and smectite are the major components of the whole  
364 glacial period, MIS3 is enriched in smectite (K/S 1/4 1.09) compared with MIS2 and MIS4  
365 (K/S 1/4 1.44 and 1.31 respectively; Table 1). The global distribution of MIS3 samples in the  
366 I/(C + K) vs I/S diagram points out a smectite-rich source (Fig. 4), characterized by a K/S  
367 ratio not corresponding to any of present-day local end-members (K/S of the Unare River 1/4  
368 1.5). This specific smectite supply may not result from changing provenance but is better  
369 explained by modifications of weathering conditions in the Unare River drainage basin during  
370 MIS3 compared to full glacial conditions. Indeed, smectite formation is favored in poorly  
371 drained soils (Wilson, 1999). Models indicate that precipitations were globally higher during  
372 MIS3 compared to LGM especially in the tropics and inter-tropical domains, associated with  
373 a higher seasonality during MIS3 compared to full glacial conditions (Van Meerbeek et al.,

374 2009). Enhanced rainfall associated with contrasted seasonal precipitations concentrated on  
375 the poorly-drained great plains area, due to the mean southward position of the ITCZ during  
376 glacials, likely favored the formation of abundant smectite. The enhanced contribution of  
377 smectite versus kaolinite thus likely reflects the combined effect of insolation and seasonality  
378 on weathering, leading to a clay association which has no present-day analog.

379 At millennial scale, latitudinal fluctuations of the ITCZ have been proposed, with a  
380 northern position of the ITCZ during interstadials and a southern position during stadials  
381 (Peterson et al., 2000a; Peterson and Haug, 2006; Deplazes et al., 2013). Nevertheless, the  
382 evolution of the clay mineral assemblages through the 20e70 ka interval (Fig. 3) does not  
383 particularly reflect changes in the source of detrital inputs. The relatively similar clay mineral  
384 assemblages in laminated and bioturbated intervals indicate that from MIS4 to the LGM, the  
385 source of clay minerals broadly remained the same and was largely dominated by inputs from  
386 the Unare River. This implies that even if the latitudinal position of the ITCZ fluctuated in  
387 relation to stadial/interstadial cycles (Peterson et al., 2000a; Peterson and Haug, 2006;  
388 Deplazes et al., 2013), the summer ITCZ remained in a relatively southward latitudinal  
389 position and never reached the drainage basin of the Tuy and Neveri rivers (Fig. 6C).  
390 Millennial scale oscillations of the K/S ratio over this interval (Fig. 3) however suggest  
391 variations in runoff intensity or erosion type, which likely altered the kaolinite-to-smectite  
392 ratio (Stern et al., 1991). The K/S ratio is low during interstadials, whereas it is maximum  
393 during stadials (Fig. 3). The higher smectite content observed in the laminated stadial  
394 sediments is coeval with an increased terrigenous fraction (Peterson et al., 2000a; Peterson  
395 and Haug, 2006; González et al., 2008a) (Figs. 3 and S4). It is consistent with increased  
396 runoff due to a northerly position of summer ITCZ and/or increased seasonality during these  
397 periods (Fig. 6C). Conversely, the higher kaolinite content during, or at the beginning of,  
398 Heinrich-like events (Fig. 3) associated to smaller grains could reflect decreased runoff and  
399 therefore more arid conditions during these periods (Peterson et al., 2000a; Peterson and  
400 Haug, 2006; González et al., 2008a) and/or could result from slight decrease of seasonality,  
401 which favor kaolinite versus smectite formation (Fig. 6C).

402 Palynological data from the Cariaco sediments suggest rapid sea-level rises during the  
403 course of Heinrich-like events (González and Dupont, 2009), promoting remobilization of  
404 sediments from the Unare platform. Clay mineral analyses of surface sediments from the  
405 Unare platform indicate that these sediments are rich in kaolinite compared to the sediment  
406 deposited at the river mouths, due to differential sedimentation (Bout-Roumazelles et al.,  
407 2013). The high kaolinite content at the beginning of Heinrich-like events (Fig. 3) is also

408 associated to a decrease of the mean grain size of the sediment (Fig. 4). This suggests that  
409 Heinrich-like events are associated to the delivery of clayey kaolinite-rich sediments eroded  
410 from the Unare platform, consistent with the global rapid sea-level rise at the beginning of  
411 Heinrich-like events (Siddall et al., 2003; Rohling et al., 2004; González and Dupont, 2009).  
412 Supporting this interpretation, several rapid sea level fluctuations are locally documented in  
413 the Gulf of Cariaco during MIS3, that were associated to Heinrich events 4e6 (Van Daele et  
414 al., 2011).

415 During MIS6, MIS5 and MIS2, some peaks in kaolinite content evidenced by changes  
416 of either K/I or K/S ratios are associated to modifications of the sedimentary characteristics  
417 (Fig. 4): a decrease of the mean grain size (Fig. 4) and/or changes in the magnetic  
418 susceptibility (Fig. S3). All these kaolinite peaks are coeval to North Atlantic cold events  
419 (Chapman and Shackleton, 1999; Lehman et al., 2002). Even if comparison with detailed sea-  
420 level variation curves (Siddall et al., 2003; Rohling et al., 2004, 2008) indicates that most  
421 peaks also correspond to periods of rapid sea level rises, their distribution in the I/S vs I/(C +  
422 K) plot is relatively different from that of Unare platform samples (Fig. S5), suggesting that  
423 these kaolinite-rich intervals do not result from sediment remobilization, but rather reflect  
424 high-latitude forcings on low-latitude climatic conditions.

425

#### 426 4.4. Younger Dryas e Termination II

427 The Younger Dryas (YD) sediments of the Cariaco Basin are characterized by a  
428 relatively high illite content and a marked drop in kaolinite content, resulting in especially  
429 high values of the I/K ratio (up to >3; Fig. 4). Moreover, the YD is characterized by a specific  
430 supply in IS (19%) in both MD02-2625 and ODP 1002 cores. As discussed for the Holocene,  
431 the high IS content suggests enhanced contribution from the Araya/Manzanares area. In the  
432 I/S vs I/(C + K) diagram, YD samples plot out of the Unare-Neveri-Tuy triangle (Fig. 5),  
433 pointing to an additional contribution by the Araya area. However the peculiar I/K ratio  
434 observed during the YD raises the question of a possible eolian contribution in the Cariaco  
435 Basin. In low latitudes where the Cariaco Basin is located, intervals of high eolian activity  
436 mostly correspond to the Younger Dryas and Heinrich-like events (deMenocal et al., 2000a;  
437 Jullien et al., 2007; Itambi et al., 2009). In particular, strong wind activity over North Africa  
438 (Lancaster et al., 2002) and high inputs of eolian material in western tropical Atlantic during  
439 the YD are well documented (deMenocal et al., 2000a; Jullien et al., 2007). Evidence for  
440 strong wind activity in Venezuela during the YD is also given by the dune fields of the  
441 Orinoco plains, where the most recent dune generation rests on a paleosol dated to 11.1 14C

442 ka (Clapperton, 1993).

443 Saharan air-borne particles recovered at Barbados are dominated by illite or kaolinite  
444 (Glaccum and Prospero, 1980; Caquineau et al., 1998, 2002). Recent data from the coast of  
445 Senegal -acquired in our laboratory with the same analytical protocol (Skonieczny et al.,  
446 2013)- allow direct comparison with the Cariaco data. The comparison of both sides of the  
447 Atlantic Ocean is moreover possible since the I/K ratio of Saharan dust remains relatively  
448 constant during the travel across the Atlantic Ocean (Glaccum and Prospero, 1980; Caquineau  
449 et al., 1998). The I/K ratio of the eolian material recovered in Senegal ranges between 0.57  
450 and 0.81, with a mean value of 0.62 (Skonieczny et al., 2013). These values are close to the  
451 average value observed in the Cariaco sediments (Fig. 4), but much lower than the values  
452 observed during the YD. The I/K ratio of Saharan dust varies according to the source of the  
453 material, and values up to 2.2 are observed in the North Western Sahara (Caquineau et al.,  
454 2002; Skonieczny et al., 2013). We cannot therefore rule out that the eolian material present  
455 in the Cariaco Basin sediments during the YD originated from high I/K areas of Northwestern  
456 Africa. However, the source of the eolian contribution needs not being located in North  
457 Africa: in the I/S vs I/(C + K) diagram, YD samples point towards the Araya-Margarita sector  
458 (Fig. 5). The Araya-Margarita sector also shows high I/K ratios (1.4° 2.2), that are in the range  
459 of the high values observed during the YD. The Araya-Margarita area is located upwind to the  
460 Cariaco Basin and currently has a mostly semi-arid climate (Fig. 6A). It appears possible that  
461 the generally windier conditions of the YD favored even drier conditions in the Araya-  
462 Margarita area, soil erosion providing material for wind transport. Overall, these data are  
463 consistent with a relatively southward position of the ITCZ during the YD, but characterized  
464 by a very peculiar wind strength or wind pattern that had no equivalent during the climatic  
465 cycle (Fig. 6B). The Preboreal interval (PB) has a clay mineral content that is intermediate  
466 between those of the YD and Holocene (Table 1, Fig. 5), suggesting that the eolian  
467 contribution did not stop immediately at the end of the YD, bur rather progressively faded  
468 during the course of the PB.

469 Though lower than for the YD, high I/K ratios are also observed for MIS6-5 transition  
470 (Fig. 4), associated with slightly enhanced IS contribution. When plotted on the I/S vs I/(C +  
471 K) diagram, samples from Termination II are shifted in the same direction as samples of the  
472 YD, possibly also indicating an eolian contribution during that period. The sediment  
473 deposited during the MIS6-5 transition somehow resembles the one deposited during the YD:  
474 light colored laminations are clearly visible and diatoms are abundant (Peterson et al., 2002).  
475 The presence of eolian material in the sediment of the MIS6-5 transition gives support to the

476 hypothesis that YD-like windy conditions, promoting upwelling activity in the Cariaco Basin,  
477 existed during this interval. A cold YD-like event at the MIS6-5 transition has been described  
478 in several terrestrial and marine records (Seidenkrantz et al., 1996; Sánchez Goñi et al., 1999;  
479 Cannariato and Kennett, 2005). This “Termination II climate oscillation” as it is sometimes  
480 referred to, was of lower intensity than the YD (Kukla et al., 2002). Consistent with this  
481 observation, the Cariaco record showing a lower value of the I/K ratio compared to the YD  
482 points to a lower eolian contribution and suggests reduced wind intensity.

483

## 484 5. Conclusion

485         Based on the recent reevaluation of the sources of clay minerals in the Cariaco Basin  
486 (Bout-Roumazielles et al., 2013), our new record of clay mineral variations in the Cariaco  
487 sediments during the last climatic cycle allows to re-consider how climatic fluctuations were  
488 recorded by the detrital flux in this basin.

489         Smectite-rich sediments from the Unare River, representing a southern detrital source,  
490 dominate the clay-mineral flux to the central Cariaco Basin throughout the studied interval.  
491 Its contribution is almost exclusive during glacial, while an increased contribution of illite-  
492 rich sediments from northern detrital sources is observed during interglacials. At orbital  
493 timescale, this change of the respective contributions of detrital sources mostly has a direct  
494 climatic origin and is linked to the mean latitudinal position of the ITCZ. Moreover, the clay  
495 composition of MIS3 suggests that the effect of insolation in driving the mean latitudinal  
496 position of the ITCZ (location of the precipitation belt, and total rainfall) is modulated by  
497 seasonality (monthly rain distribution), which modified the weathering regime of the drainage  
498 basin.

499         Rapid fluctuations of the clay mineral content during the Last Glacial are related to  
500 millennial Dansgaard-Oeschger oscillations and Heinrich-like events. These oscillations are  
501 likely linked with fluctuations of runoff intensity (Peterson et al., 2000a; Peterson and Haug,  
502 2006), but the clay-mineral association rules out any significant change of the origin of the  
503 detrital flux. The observed variations of the clay-mineral content rather respond to slight  
504 increases of seasonality during interstadials while decreased seasonality and remobilization of  
505 sediment from the Unare platform associated to rapid sea level variations characterized  
506 Heinrich-like events and stadials.

507         The clay data clearly point for a significant eolian contribution to the Cariaco Basin  
508 during the Younger Dryas, confirming that this period was marked by high wind intensity in  
509 the subtropical belt. According to its clay-mineral content, this eolian material may originate



510 from the Sahara and/or from the arid Araya-Margarita area. A similar eolian contribution is  
511 also observed at the MIS6-5 transition pointing to the occurrence of a windy Younger Dryas-  
512 like episode during Termination II.

513

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524

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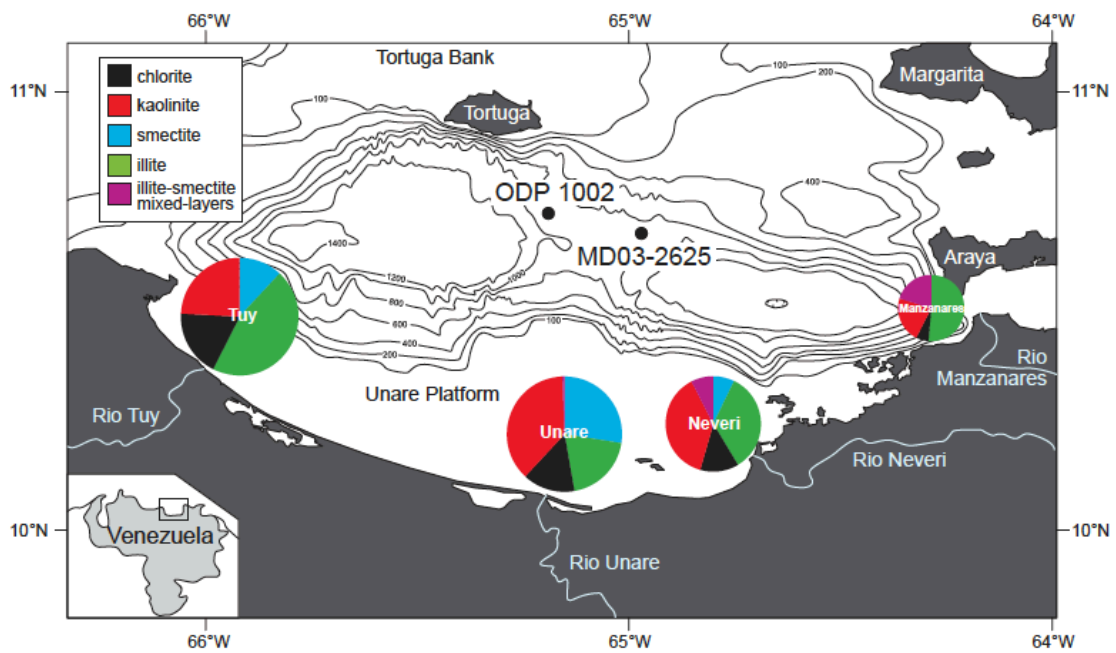
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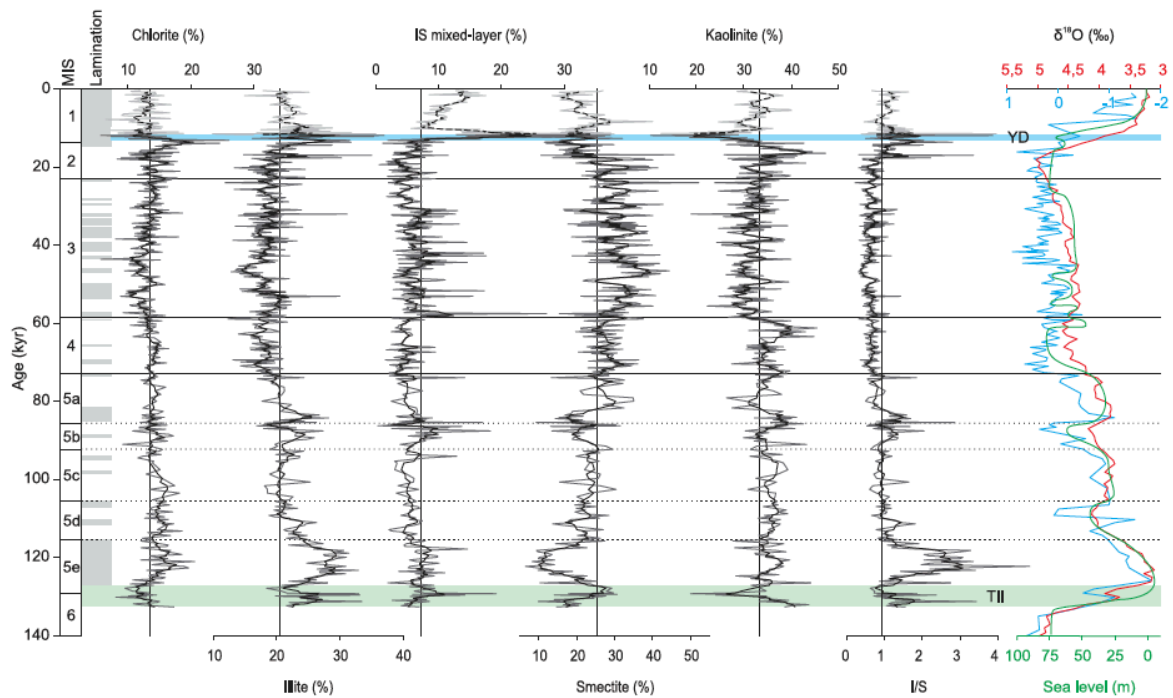


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788

789 **Figures Caption**



790  
791 Fig. 1. Bathymetry of the Cariaco Basin and location of the cores used in this study. Average clay  
792 distribution at river mouths from Bout-Roumazelles et al. (2013).



793

794 Fig. 2. Clay mineral variations in the sediments of the Cariaco Basin and illite/smectite ratio (I/S).

795 Black curves correspond to 5 points running mean. The vertical line indicates the average value. Light

796 gray curve: MD03-2625; dark gray curve: ODP 1002 hole. Intervals of sediment lamination are

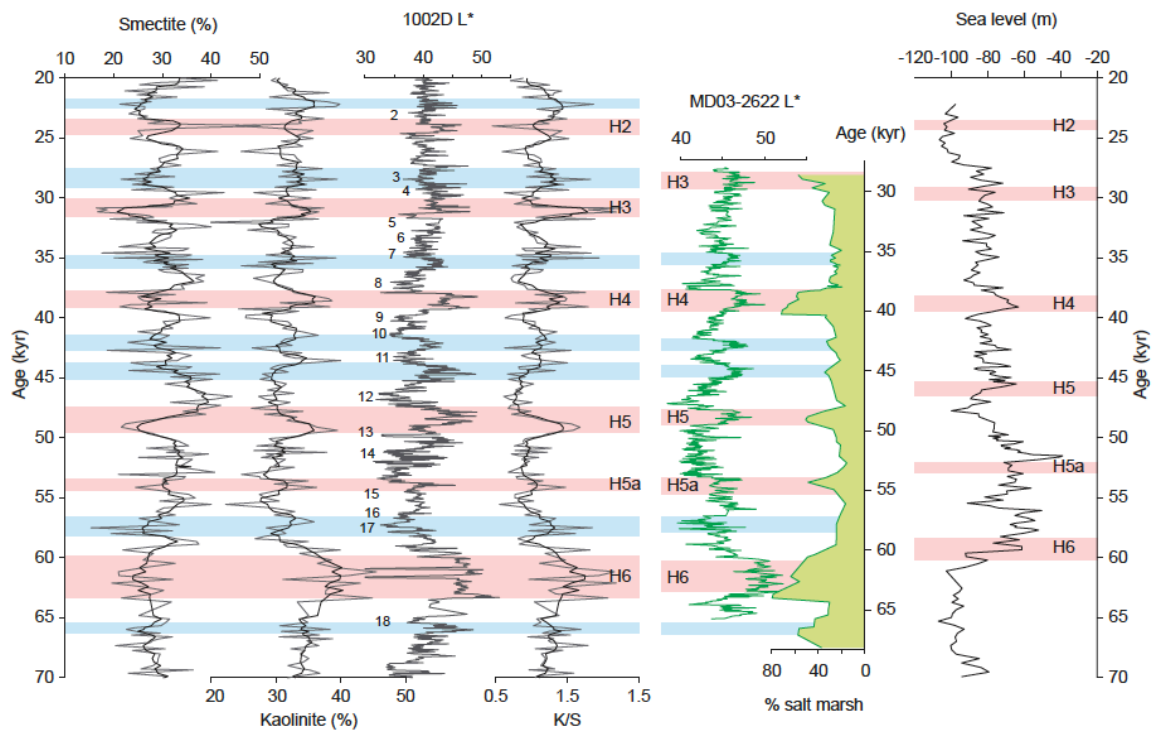
797 indicated on the left. Benthic  $\delta^{18}\text{O}$  values (red curve) from Lisiecki and Raymo (2005). Planktonic

798 values (blue curve) modified from Drenzek (2007). Gulf of Cariaco sea level variations (green curve)

799 modified from Van Daele et al. (2011). YD: Younger Dryas; TII: Termination II. (For interpretation of

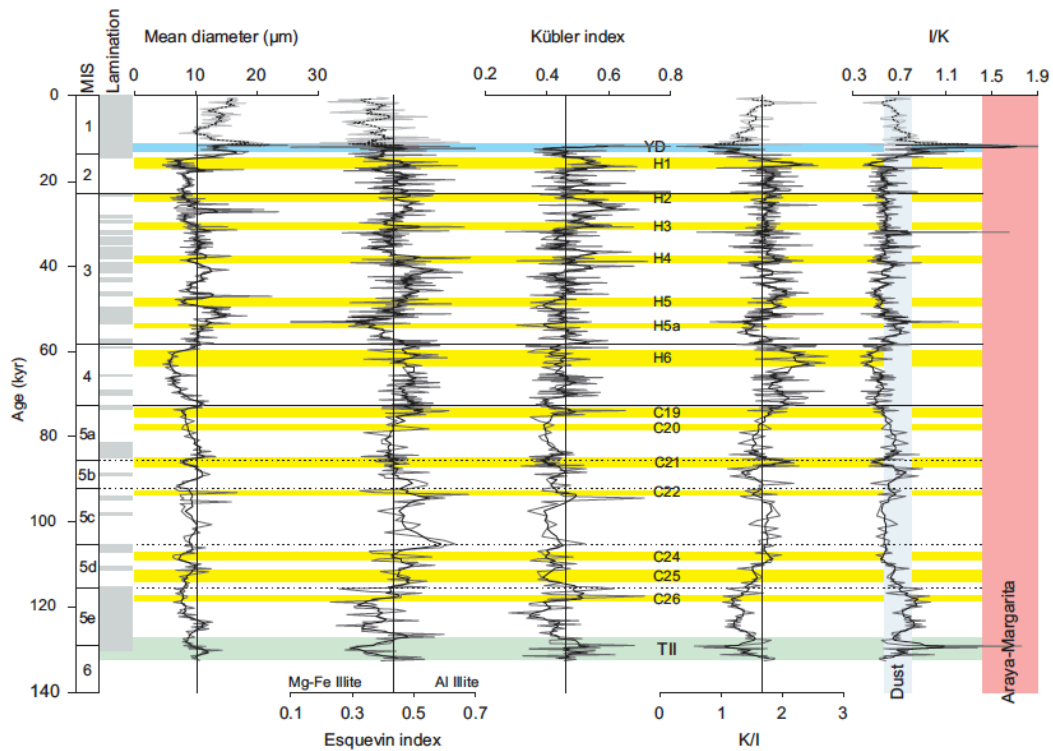
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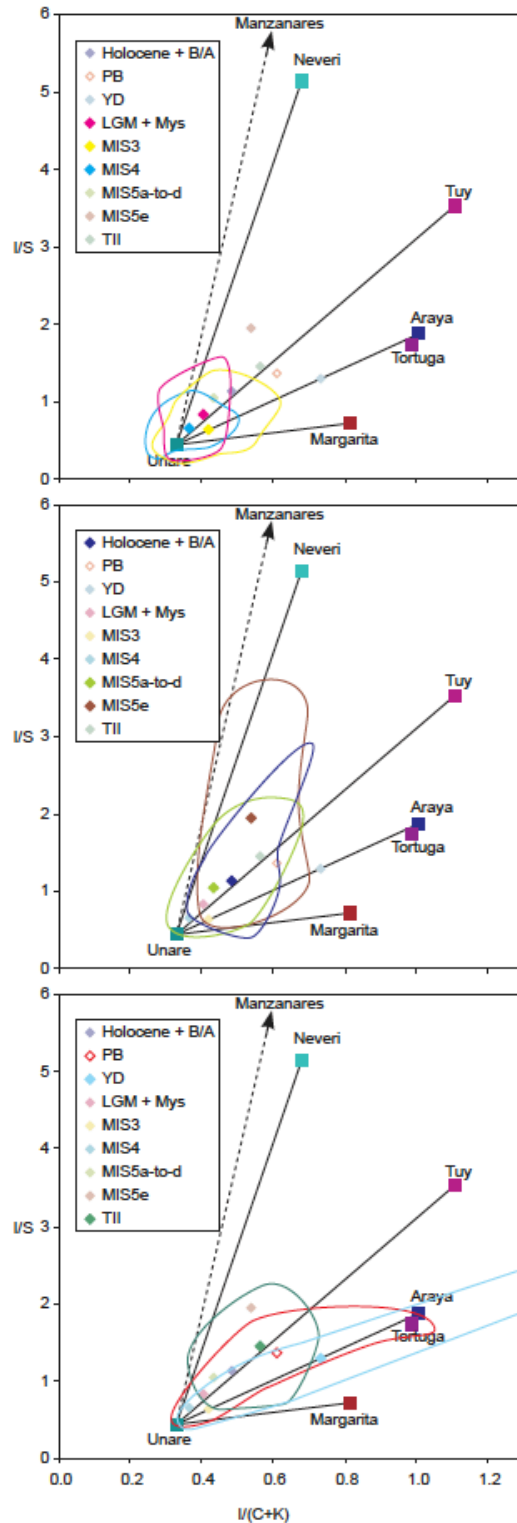
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804 Fig. 3. Detailed variations of the smectite and kaolinite contents and the kaolinite/smectite ratio (K/S)  
 805 for the 20-70 ka interval. Black curves correspond to 5 points running mean. Sediment reflectance  
 806 (L\*) for ODP 1002 D hole modified from Shipboard Scientific Party (1997). These data are compared  
 807 to the record of salt marsh pollens and sediment reflectance in core MD03-2622 from the Cariaco  
 808 Basin (González and Dupont, 2009), and detailed sea level variations in the Red Sea (Siddall et al.,  
 809 2003). Notice that different age models were used for the three records. Numbers of  
 810 Dansgaard/Oeschger stadials are indicated on the left side of the 1002D reflectance curve (Peterson et  
 811 al., 2000a, 2000b). Levels in pink indicate Heinrich-like events. Levels in blue underline the good  
 812 consistency in the clay mineral and palynological records. (For interpretation of the references to  
 813 colour in this figure legend, the reader is referred to the web version of this article.)



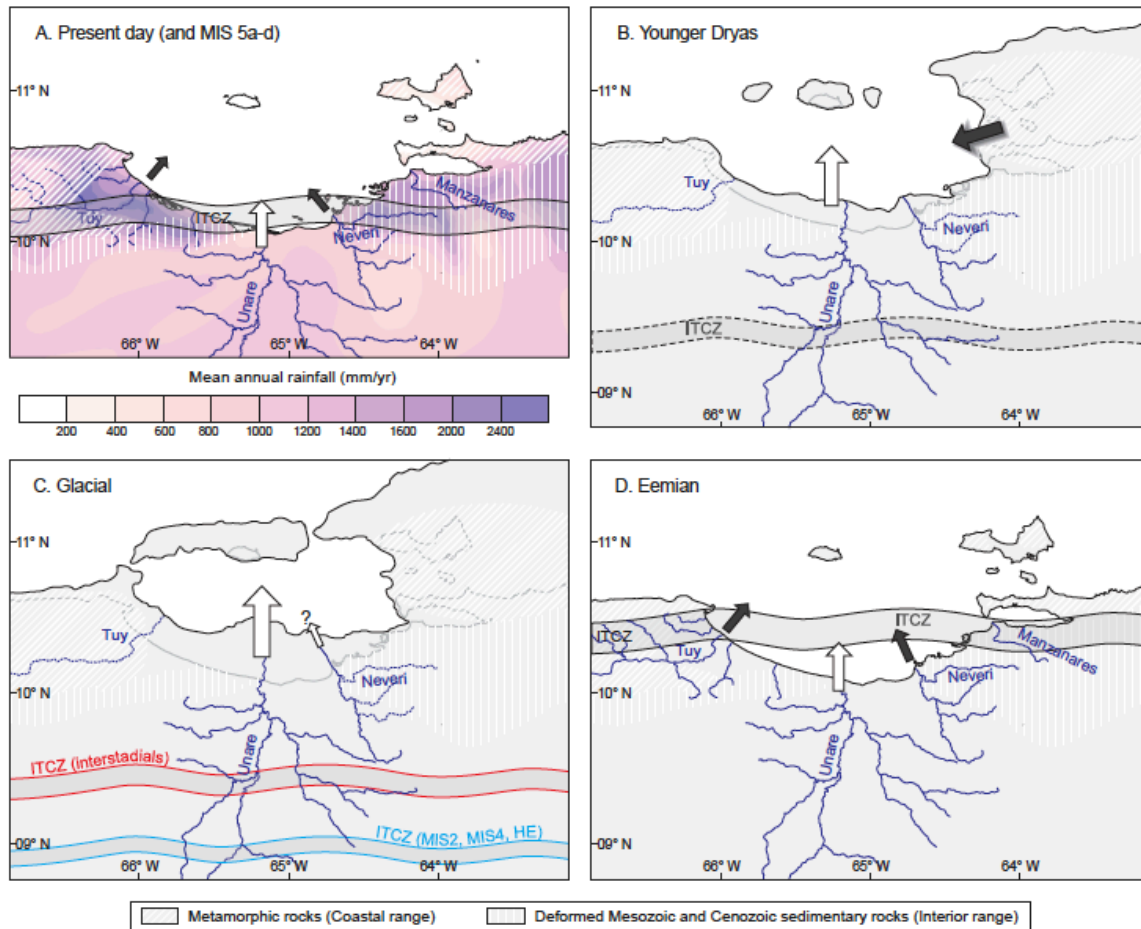
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815 Fig. 4. Variations of the decarbonated sediment mean grain size, Esquevin index, Kübler index,  
 816 kaolinite/illite ratio (K/I) and illite/kaolinite ratio (I/K) in the sediments of the Cariaco Basin. Black  
 817 curves correspond to 5 points running mean. Light gray curve: MD03-2625; dark gray curve: ODP  
 818 1002 hole. Vertical blue band: range of values of the I/K ratio in aerosols collected in Senegal  
 819 (Skonieczny et al., 2013). Vertical pink band: range of values in the platform sediments from the  
 820 Araya-Margarita sector (Bout-Roumazelles et al., 2013). Intervals of sediment lamination are  
 821 indicated on the left. Yellow horizontal bands indicate Heinrich-like events and cold stadials. YD:  
 822 Younger Dryas; TII: Termination II. (For interpretation of the references to colour in this figure  
 823 legend, the reader is referred to the web version of this article.)



824

825 Fig. 5. Binary diagram illite/smectite ratio (I/S) versus illite/(chlorite + kaolinite) ratio (I/(C + K))  
 826 showing the area of distribution and average values of Cariaco Basin sediments from different  
 827 intervals compared to the average values of the different detrital sources around the basin (Bout-  
 828 Roumazelles et al., 2013). Areas of distribution of different time intervals are highlighted in the  
 829 different panels for clarity. B/A: Bølling- Allerød; PB: Preboreal; YD: Younger Dryas; LGM - Mys:  
 830 Last Glacial Maximum to “Mystery interval” (26.5e14.6 ka). MIS: marine isotopic stage.



831  
 832 Fig. 6. Simplified geological map of North Venezuela (modified from Hackley et al., 2006) and  
 833 schematic diagrams comparing the sources and transport of clay minerals to the Cariaco Basin and  
 834 position of the summer ITCZ for different time intervals. A. Present-day geography and pluviosity  
 835 map (from Goldbrunner, 1984), schematic position of summer ITCZ for the present and MIS5a-to-d.  
 836 B. Younger Dryas. The position of summer ITCZ is uncertain. The shaded arrow indicates eolian  
 837 contribution. C. Last Glacial Maximum geography and schematic position of the ITCZ during  
 838 Heinrich events, MIS2 and MIS4 (blue) and during interstadials (red). D. Eemian. White arrows  
 839 represent smectite-rich sediments. Dark gray arrows illite-rich sediments. The size of the arrows  
 840 represents the respective contribution to the detrital flux. The width of the ITCZ band increases with  
 841 seasonality. (For interpretation of the references to colour in this figure legend, the reader is referred to  
 842 the web version of this article.)  
 843

844 Table 1

845 Average clay mineral content and ratios of the Cariaco sediments for different time intervals.

	Chlorite %	Illite %	IS %	Smectite %	Kaolinite %	I/S	I/(C + K)	K/S
Holocene + B/A	13.9	22.2	10.3	21.2	32.3	1.14	0.49	1.65
Preboreal <sup>a</sup>	12.5	23.4	17.8	19.4	26.9	1.36	0.61	1.45
Younger Dryas	12.2	23.2	19.4	21.4	23.8	1.29	0.73	1.19
LGM + Mys <sup>b</sup>	14.1	19.4	5.0	26.9	34.6	0.82	0.40	1.44
MIS3	13.0	18.4	6.4	30.7	31.5	0.64	0.42	1.09
MIS4	13.8	17.9	4.8	28.1	35.4	0.66	0.37	1.31
MIS5a-to-d	14.3	21.6	5.8	22.5	35.7	1.04	0.43	1.69
MIS5e	14.7	26.5	7.3	16.7	34.8	1.94	0.54	2.51
TII	12.8	25.4	7.5	19.9	34.4	1.44	0.56	1.89

846

847 b Last Glacial Maximum and “Mystery interval”: from 26.5 to 14.6 ka.

848

849 Table 2

850 Average contribution (%) of the Tuy, Unare and Neveri Rivers as determined from a simple mixing  
851 model using Fig. 5.

	Tuy	Neveri	Unare
Holocene + B/A	19	2	79
LGM + Mys	8	4	88
MIS4	4	2	94
MIS5ad	11	6	83
MIS5a	12	5	83
MIS5b	12	5	83
MIS5c	11	3	86
MIS5d	10	7	83
MIS5e	18	20	62
TII	26	3	71

852