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► To cite this version:

Pauline Corentin, Emmanuelle Pucéat, Pierre Pellenard, Michel Guiraud, Justine Blondet, et al.. Late Cretaceous evolution of chemical weathering at the northeastern South American margin inferred from mineralogy and Hf-Nd isotopes.. *Marine Geology*, 2023, 455, pp.106968. 10.1016/j.margeo.2022.106968 . hal-03911852

HAL Id: hal-03911852

<https://u-bourgogne.hal.science/hal-03911852>

Submitted on 23 Feb 2024

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1 **Late Cretaceous evolution of chemical weathering at the northeastern South American**
2 **margin inferred from mineralogy and Hf-Nd isotopes**

3

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11

12 *Abstract*

13 The origin of the global temperature decrease that initiated the last greenhouse-icehouse
14 transition about 90 million years ago still remains debated. Among the potential processes dri-
15 ving long-term climate evolution over million-year timescales, this study investigates the
16 possible impact of the uplift-weathering connection on the late Cretaceous atmospheric CO₂
17 drawdown and associated cooling.

18 We analysed a marine sediment record from the Demerara margin, using Nd and Hf
19 isotopes in clay-size detrital fractions ($\Delta\epsilon_{\text{Hf}(t)\text{clay}}$) together with bulk and clay mineralogy and
20 major element abundances to reconstruct the evolution of tectonic uplift and chemical
21 weathering intensity in northeastern South America during the late Cretaceous. Our data
22 indicate that silicate weathering intensified on the northeastern South American margin during
23 the middle Campanian and the Maastrichtian, concomitant with an uplift phase of the Guiana
24 craton. We propose that the tectonic pulse highlighted by apatite fission track data,

25 argon/argon dating and primary silicate mineral evolution, was accompanied by an
26 accelerated chemical weathering, which presumably acted as a sink for atmospheric CO₂. By
27 contrast, during the earlier late Turonian to early Campanian, period of relative tectonic
28 quiescence, climate most likely acted as the main driver controlling the evolution of chemical
29 weathering intensity. The main phase of enhanced weathering induced by the uplift of the
30 northeastern South American margin occurred during the Campanian, thereby postdating the
31 onset of global seawater temperature decrease (late Turonian). By inference, this regional
32 event is unlikely to have triggered late Cretaceous cooling but it may have acted as an addi-
33 tional factor stabilizing colder climate conditions at that time.

34

35 *1. Introduction*

36 The late Cretaceous was a period marked by major tectonic and climatic changes. While a
37 global decrease of seawater temperature from the Turonian to the Maastrichtian is recorded by
38 Tex₈₆ and foraminiferal δ¹⁸O proxy data at all latitudes, its origin remains unclear (Friedrich
39 *et al.*, 2012 ; O'Brien *et al.*, 2017). A pronounced decrease in atmospheric CO₂ levels (*p*CO₂)
40 at that time (Wang *et al.*, 2014) indicates that the late Cretaceous climate cooling was closely
41 related to the global long-term carbon cycle. The same period was also characterized by major
42 tectonic events such as the uplift of the West African and eastern South American margins
43 (Harmann *et al.*, 1998 ; Gallagher & Brown, 1999 ; Jaspert *et al.*, 2012 ; Engelmann de
44 Oliveira *et al.*, 2016 ; Green *et al.*, 2017, 2018 ; Murlot *et al.*, 2018 ; Roddaz *et al.*, 2021),
45 which could have impacted the carbon cycle through enhanced mechanical erosion and
46 chemical weathering.

47 Indeed, among the processes driving the long-term carbon cycle, chemical weathering of
48 silicate rocks on continents acts as a major sink of atmospheric CO₂ over million-year

49 timescales. Chemical weathering depends on climatic parameters such as temperature and
50 precipitation (White & Blum, 1995), but also on mechanical erosion rates. Continuous
51 exposure of fresh rock surfaces by mechanical erosion promotes chemical weathering (Raymo
52 & Ruddiman, 1988 ; West *et al.*, 2005 ; Gabet & Mudd, 2009), up to a threshold where, at
53 high denudation rates, detrital material is exported too rapidly to further sustain silicate
54 weathering in soils (West *et al.*, 2005 ; Gabet & Mudd, 2009). In active tectonic settings, such
55 kinetically-limited weathering regimes typically lead to near-constant chemical weathering
56 rates with increasing denudation rates (White & Blum, 1995 ; Gabet & Mudd, 2009 ; West,
57 2012). Therefore, the relationship between tectonic uplift and silicate weathering, and thus the
58 net impact of this process on atmospheric carbon concentrations, is not straightforward and
59 still needs to be explored. The major uplift phase of South Atlantic margins during the late
60 Cretaceous major climate change could provide new insights to explore the interplay between
61 climate and tectonic uplift.

62 In this study, we investigated a marine sediment record from Demerara Rise (ODP Site
63 1259) to explore the response of silicate weathering to the uplift of the northeastern margin of
64 the South American continent, which occurred during the late Cretaceous from Guiana to
65 Brazil (McConnell, 1968 ; Harman, 1998 ; Morais Neto *et al.*, 2009 ; Jaspen *et al.*, 2012 ;
66 Green *et al.*, 2018). A multi-proxy approach based on Hf-Nd isotope analyses of clay-size
67 detrital fractions combined with complementary geochemical and mineralogical data can
68 provide unique insights into the evolution of continental chemical weathering through time
69 (Bayon *et al.*, 2016 ; Coarentin *et al.*, 2022; Bayon *et al.*, 2022). Recently, the same approach
70 has been successfully used to reconstruct the late Cretaceous evolution of silicate weathering
71 along the southeastern Brazilian margin (Coarentin *et al.*, 2022), showing that the formation of
72 high topographic relief under arid climate conditions favoured mechanical erosion until the

73 elevation was high enough to support high orographic precipitations and, consequently,
74 enhanced silicate chemical weathering (Corentin *et al.*, 2022).

75

76 2. Setting

77 ODP Site 1259 (2354 meter depth) from Demerara Rise (Fig. 1) exhibits upper Cretaceous
78 sediments from the Cenomanian to the Maastrichtian (Erbacher *et al.*, 2004). The samples
79 were collected from two lithostratigraphic units (between 440 and 540 mbsf; Fig. 2, 3) : 1) a
80 Turonian to Santonian interval composed of laminated calcareous claystone with organic
81 matter, clayey chalk and limestone; and 2) a Campanian to late Maastrichtian interval,
82 composed of nannofossil-rich clayey chalk, calcareous siltstone and glauconitic claystone
83 (Fig. 2, 3 ; Supplementary Table 1 ; Erbacher *et al.*, 2004). These sediments were deposited in
84 a marine setting evolving from shelf environments during the Cenomanian to upper bathyal
85 environments in the early Campanian (Erbacher *et al.*, 2005), under direct influence of
86 terrigenous fluxes from the adjacent continental margin (März *et al.*, 2009).

87 The Demerara Rise, surrounded by the Guiana/Suriname (GS) and the Foz do Amazonas
88 (FOZ) basins, is located on the northeastern South American margin (Fig. 1.A, 1.B), near the
89 Guiana, Suriname and French Guiana coastal regions (Fig. 1.B). During the end of the early
90 Cretaceous, a plate kinematic reorganization (~ 105 Ma), followed by a late Cenomanian-
91 Turonian deformation led to the uplift of the Demerara plateau (Yang and Escalona, 2011;
92 Basile *et al.*, 2013; Loncke *et al.*, 2020). The plateau became a structural high separating the
93 Guiana/Suriname Basin and the Foz do Amazonas Basin and resulting in the isolation the
94 Demerara plateau (Fig. 1.B; Loncke *et al.*, 2020). After the late Albian unconformity, the
95 Demerara plateau evolved into a passive margin with a transition from shallow to deep marine
96 conditions (Loncke *et al.*, 2020).

97 The Guiana Shield represents the main continental landmass that provides detrital sediment
98 inputs to the Demerara Rise (Roddaz *et al.*, 2021 ; Fig. 1.B). During the late Cretaceous, this
99 area corresponded to a low-elevation passive margin located in a humid equatorial climatic
100 belt (Hay *et al.*, 2012 ; Sapin *et al.*, 2016).

101

102 3. Material and methods

103 A total of 88 samples from Site 1259 were analysed, spanning a period from the late Tu-
104 ronian to late Maastrichtian. The age model is based on the biostratigraphy published in
105 Erbacher *et al.* (2004) and Gradstein *et al.* (2012).

106 3.1. Mineralogical analyses

107 Bulk sediment and clay (< 2 µm) mineralogical analyses were performed at the Biogeos-
108 ciences Laboratory (University of Burgundy, France) using X-ray diffraction (XRD) with a
109 Bruker Endeavor D4 diffractometer equipped with a Lynxeye detector, CuK α radiations and
110 Ni filter, under 40 kV voltage and 25 mA intensity. The isolated clay fraction was analysed
111 using the three treatments recommended by Moore and Reynolds (1997). Identification and
112 semi-quantification of the mineral phases were performed with the software MacDiff 4.2.5
113 (Petschick, 2001) using the position of mean diffraction peaks and the area of their main
114 diffraction peaks, respectively. For clay mineralogy, the proportions were estimated after
115 ethylene glycol solvated preparation using the main diffraction (d_{001}) peak.

116 The (Illite+Chlorite)/smectite ((I+C)/S), (Opal+Clinoptilolite)/Calcite ((O+C)/Ca) and
117 Feldspar/Clays (Feld./Clays) ratios were calculated using the mean peak area of each mineral.
118 The uncertainty of the relative proportions of clay minerals is estimated as < 5%, based on
119 replicate analyses of samples and the internal procedure of quantification using MacDiff on
120 mixtures of clay minerals in known proportions.

121 The clay fractions of 3 samples were dispersed in distilled water and butylamine solution,
122 and deposited on a carbon formvar Cu grid for Transmission Electron Microscopy (TEM) ob-
123 servations. Images of clay particles were performed using a MET Hitachi 4800 at the INRAE
124 of the University of Burgundy (France).

125 3.2. Geochemical analyses

126 Major element abundances and CIA

127 X-ray fluorescence spectroscopy (XRF) was used to determine whole-rock major element
128 abundances (SiO₂, TiO₂, Al₂O₃, Fe₂O₃, MnO, MgO, CaO, Na₂O, K₂O and P₂O₅;
129 Supplementary Table 1) at the University of Lausanne, using a wavelength-dispersive
130 PANalytical AxiosmAX spectrometer fitted with a 4 kW Rh X-ray tube. Fused disks were
131 prepared from 1.2 g of calcined sample powder mixed with lithium tetraborate (1:5 mixture)
132 for the analyses. A number of 21 international silicate rock reference materials was used for
133 the calibration. The data are reported on a loss of ignition (LOI)-free basis (Supplementary
134 Table 1). The limits of detection are in the range 20 to 80 ppm for major elements depending
135 on the element concerned.

136 The chemical index of alteration (CIA) was calculated and used as a proxy of chemical
137 weathering intensity (Nesbitt & Young, 1982). Because our samples of bulk sediments
138 display relatively high carbonate contents (between 14-91 %), detrital CaO contents were
139 estimated using the method of McLennan (1993), which consists of replacing CaO* by Na₂O
140 in the following calculation: $(Al_2O_3/[Al_2O_3 + Na_2O + CaO^* + K_2O]) \times 100$ (where CaO*
141 represents the amount of CaO in the silicate fraction of the sediments). The CIA is generally
142 interpreted as the extent to which feldspar, which contains relatively mobile Ca, Na and K,
143 has been weathered and converted to Al-rich clay minerals (Nesbitt & Young, 1982).

144 Hf, Zr and REE concentrations and Hf-Nd isotopes

145 Following the protocol described in Bayon *et al.* (2002) and modified in Coentim *et al.*
146 (2022), a total of 23 samples (8 samples collected in claystone lithology and 15 samples
147 collected in chalk lithology ; Supplementary Table 1) were selected for geochemical analyses.
148 This protocol includes a sequential leaching procedure that successively removes carbonates,
149 Fe-Mn oxyhydroxides and organic matter. Then, the clay-size detrital fraction ($< 2\mu\text{m}$) is
150 separated from the residual sediments by decantation and submitted to a second leaching
151 procedure. Following clay extraction, about 65 mg of dry powdered samples were digested by
152 alkaline fusion after addition of a Tm spike (Barrat *et al.*, 1996).

153 Finally, ion exchange chromatography was used to isolate purified Nd and Hf fractions
154 using columns packed with AG-50W-X8 and Ln-Spec resin for Nd and AG1-X8 and Ln-Spec
155 for Hf, following the protocols described in Chu *et al.* (2002) and Bayon *et al.* (2012), res-
156 pectively.

157 Concentrations of REE, Hf and Zr were measured by inductively coupled plasma mass
158 spectrometry (ICP-MS) on a Thermo Scientific X-Series II at the Pôle Spectrométrie Océan
159 (Brest, France). Details about measured elemental masses, polyatomic interference
160 corrections and the quantification method are given in Barrat *et al.* (1996) and Bayon *et al.*
161 (2009b). External reproducibility and accuracy were estimated by replicate analyses of four
162 certified reference materials from the United States Geological Survey (BCR-2, BIR-1, MAG-
163 1, BHVO-2 ; Supplementary Table 2). Analytical reproducibility was better than 2% for all
164 elements. Deviations from the reference values for BHVO-2, BCR-2 and BIR-1 (Jochum *et*
165 *al.*, 2016) were always $< 10\%$ RSD.

166 Hafnium and neodymium isotope compositions of studied samples were measured on a
167 MC-ICP-MS Neptune Plus (Thermo Scientific) at the ENS of Lyon (France) with a sample-
168 standard bracketing method, using concentration-matched solutions of JMC475 and JNdi-1
169 standards, respectively. The instrumental mass bias were corrected applying an exponential

170 law, using $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$ and $^{179}\text{Hf}/^{177}\text{Hf} = 0.7325$. Nd and Hf procedural blanks were
171 respectively better than 6 pg and 10 pg.

172 The analytical session gave a $^{176}\text{Hf}/^{177}\text{Hf}$ ratio of 0.282163 ± 0.000008 (2 s.d., n=21)
173 corresponding to an external reproducibility of $\pm 0.28\epsilon$ (2 s.d.) based on repeated analyses of
174 JMC 475 standard solutions (at 35, 30 or 20 ppb). Replicate analyses of the BHVO-2
175 reference material gave a $^{176}\text{Hf}/^{177}\text{Hf}$ value of 0.283108 ± 0.000005 (2 s.d., n=3), in
176 agreement with the reference values of 0.283096 ± 0.000020 (Weis *et al.*, 2005).

177 The analytical session gave a $^{143}\text{Nd}/^{144}\text{Nd}$ ratio of 0.512115 ± 0.000007 (2 s.d., n=14),
178 corresponding to an external reproducibility of $\pm 0.14 \epsilon$ (2 s.d.) based on repeated analyses of
179 bracketed JNdi-1 standard solution (at 40 ppb). Repeated analyses of the BHVO-2 and BCR-2
180 reference materials gave respectively a $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.512984 ± 0.000006 (2 s.d., n =
181 3) and 0.512643 ± 0.000005 (2 s.d., n = 3), respectively, agreeing well with published values
182 of 0.512990 ± 0.000010 and 0.512638 ± 0.000015 (Weis *et al.*, 2006).

183 The Hf isotope data are reported using the standard notation $\epsilon_{\text{Hf}(t)} =$
184 $[\left(\frac{^{176}\text{Hf}/^{177}\text{Hf}}{(^{176}\text{Hf}/^{177}\text{Hf})_{\text{CHUR}}}\right) - 1] \times 10^4$, corrected for the radioactive decay of ^{176}Lu
185 to ^{177}Hf based on Hf and Lu concentrations measured for each sample ($^{176}\text{Lu}/^{177}\text{Hf} = \text{Lu}/\text{Hf} \times$
186 0.1424), the absolute age (t) calculated using the age model of Erbacher *et al.* (2004) and the
187 timescale of Gradstein *et al.* (2012), and the ^{176}Lu radioactive decay constant λ (1.867×10^{-11}
188 an^{-1} ; Söderlund *et al.*, 2004). The initial $^{176}\text{Hf}/^{177}\text{Hf}$ ratio of the CHondritic Uniform
189 Reservoir was calculated using the present-day CHUR value of 0.282785 and a $^{176}\text{Lu}/^{177}\text{Hf}$
190 ratio of 0.0336 (Bouvier *et al.*, 2008).

191 The Nd isotope data are reported using the standard notation $\epsilon_{\text{Nd}(t)} =$
192 $[\left(\frac{^{143}\text{Nd}/^{144}\text{Nd}}{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}}\right) - 1] \times 10^4$ corrected for the radioactive decay of
193 ^{147}Sm to ^{143}Nd based on measured Nd and Sm concentrations ($^{147}\text{Sm}/^{144}\text{Nd} = \text{Sm}/\text{Nd} \times$
194 0.6049), the absolute age (t) and the ^{147}Sm radioactive decay constant λ ($6.54 \times 10^{-12} \text{an}^{-1}$;

195 Lugmair & Marti, 1977). The $^{143}\text{Nd}/^{144}\text{Nd}$ ratio of CHUR was calculated using the present-
196 day value of 0.512630 and a $^{147}\text{Sm}/^{144}\text{Nd}$ ratio of 0.1960 (Bouvier *et al.*, 2008).

197 4. Results

198 4.1. Bulk and clay mineralogy

199 The bulk sediment assemblage (Fig. 2 ; Supplementary Table 1) is composed of clays (0 to
200 36%), quartz (0.5 to 28%), K-feldspar (0 to 5%), calcite (14 to 98 %), plagioclase (0 to 11%),
201 opal-CT (0 to 58 %), clinoptilolite (0.6 to 19 %), pyrite (0 to 5 %), gypsum (0 to 10 %) and
202 traces of dolomite (≤ 1 %). From the Turonian to Santonian, a rich assemblage is composed of
203 all the identified minerals. After the Santonian, the proportions of clinoptilolite, pyrite,
204 gypsum and dolomite decrease and these minerals progressively disappeared (Fig. 2). The
205 main mineralogical evolution concerns variations in the carbonate and siliceous mineral
206 content. A net increase of the opal-CT and low-quartz content is indeed observed in the
207 Santonian to Campanian interval balanced by an apparent decrease of calcite following an
208 episode marked by an increase in clinoptilolite (Fig. 2). The plagioclase and K-feldspar
209 proportions also present a slight increase from the Coniacian to the Campanian, then record a
210 decrease from the Campanian to the Maastrichtian as well as the quartz content while the
211 proportions of clay slightly increased (Fig. 2).

212 Clay mineralogy data show that clay-size fractions at Site 1259 are dominated by smectite
213 (~85 to 100 %), with minor amounts of illite and palygorskite (both up to ~8 %), and traces of
214 kaolinite and chlorite (Fig. 3 ; Supplementary Table 1). Note that chlorite contents are
215 negligible (<1 %). Clay mineral assemblages do not present major changes along the studied
216 interval. The Turonian samples present the highest proportions of illite and palygorskite, up to
217 8%, which then decrease and remain lower than 5% from the Coniacian to the Maastrichtian
218 apart from a slight increase in illite during the Coniacian-Campanian interval (Fig. 3).

219

220 4.2. Major elements

221 Major elements were measured in the bulk sediments and gave a wide range of concentrations
222 (Table 1). The elements content vary from 1.70 to 59.93 % (mean 21.90 %) for SiO₂, from
223 0.48 to 8.11 % (mean 3.83 %) for Al₂O₃ and from 0.02 to 0.33 % (mean 0.16 %) for TiO₂.
224 The K₂O content vary from 0.01 to 1.62 % (mean 0.44 %), the CaO content from 12.37 to
225 52.91 % (mean 31.71 %) and the Na₂O content from 0.03 to 1.63 % (mean 0.82 %). Finally,
226 the content vary from 0.03 to 1.59 % (mean 0.396 %) for P₂O₅, from 0.16 % to 4.30 % (mean
227 1.33 %) for Fe₂O₃, from 0.01 % to 0.08 % (mean 0.02 %) for MnO, from 0.47 % to 1.46 %
228 (mean 0.76 %) for MgO and from 0 % to 0.03 % (mean 0.01 %) for the Cr₂O₃.

229

230 4.3. Hf, Zr and REE concentrations

231 The rare earth and trace elements concentrations are reported in Table 1. The analysed clay
232 fraction displays zirconium (Zr) concentrations between 42 and 157 ppm that are in the range
233 of modern zircon-poor clay-size fraction (Bayon et al., 2016). Rare Earth Element
234 concentrations of the samples (Table 1) have been normalized to Post-Archean average
235 Australian Shale (PAAS; Taylor et McLennan, 1985), with REE patterns displayed on Fig. 4.
236 Most of the samples present a relatively flat pattern as expected for detrital clays (Fig. 4), that
237 is also highlighted by their position in the “flat” REE pattern domain in a crossplot (Fig.5B)
238 MREE/MREE* (MREE : Middle Rare Earth Elements) versus HREE/LREE (Heavy Rare
239 Earth Elements/Light Rare Earth Elements), except for three samples (at 494.26 m, 494.74 m
240 and 506.05 m) that display a slight enrichment in HREE and a slight negative cerium anomaly
241 (Ce/Ce*) between about 0.5 and 0.7). Figure 5.A presents a crossplot HREE vs Ce/Ce*
242 highlighting the position of these three samples compared to the remaining of the samples

243

244 4.4. Hf-Nd isotopes

245 The $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$ ratios and the $\varepsilon_{\text{Nd}(t)}$ and $\varepsilon_{\text{Hf}(t)}$ values are provided in Table
246 2. The Nd isotope composition of studied samples displays unradiogenic values from -13.8 in
247 the Turonian to -15.7 in the Campanian, reaching -17.5 in the late Campanian following a
248 positive excursion with values up to -12.3 (Figs. 3, 6). A slight increase is observed during the
249 Maastrichtian with values up to -15.6. The Hf isotope composition of the studied samples
250 displays unradiogenic values between -12.2 to -6.0 (Fig. 3, 6). They first decrease from values
251 of about -7.4 during the Turonian to values of about -12.2 in the Coniacian, then increase
252 from the Campanian to the end of the Maastrichtian where they reach values of about -6.1.
253 Finally, calculated $\Delta\varepsilon_{\text{Hf}(t)\text{clay}}$ values at Demerara Rise decrease from the end of the Turonian to
254 the Santonian – earliest Campanian, from -1.3 to -5.1 (Fig. 3). Then, $\Delta\varepsilon_{\text{Hf}(t)\text{clay}}$ values increase
255 during the Campanian until the end of Maastrichtian with values up to - 0.1 (Fig. 3).

256

257 5. Discussion

258 5.1. Influence of authigenesis and diagenesis on mineralogical and geochemical signals

259 Clay mineral assemblages at Site 1259 are dominated by smectite, what is a common
260 feature of upper Cretaceous marine sediment sequences, which were shown to be mainly of
261 detrital origin (Robert, 1981 ; Chamley, 1989). This is further corroborated by TEM
262 observations (Fig. 7.B), which reveal the presence of flaky shape smectite particles, with no
263 evidence of lath-type overgrowths characteristic of clay authigenesis (Clauer *et al.*, 1990).
264 However, three samples at 494.26 m, 494.74 m and 506.05 m (80.25, 83.36 and 89.15 Ma) are
265 marked by a slight negative cerium anomaly, down to 0.53, and slight enrichments in heavy

266 rare earth elements, (Figs. 4, 5) which could possibly indicate the presence of a seawater
267 signal associated with authigenic clay formation (Fagel, 1992 ; Figs. 4, 5).

268 Interestingly, these three samples presenting some evidence of clay authigenesis formation,
269 although limited, also yield more radiogenic (less negative) ϵ_{Nd} values (Fig. 3, 8 ; Table 2).

270 At Demerara Rise, $\epsilon_{Nd(t)}$ seawater values inferred from fish tooth ϵ_{Nd} are quite negative
271 during the late Cretaceous, around -16 on average over most of the interval although
272 excursions toward more radiogenic values have been recorded during Oceanic Anoxic Event
273 2 (OAE2, Cenomanian-Turonian), up to -8, and at the end of the Maastrichtian (Fig. 8;
274 MacLeod et al., 2008; Martin et al., 2012). Although the origin of the radiogenic excursion of
275 seawater $\epsilon_{Nd(t)}$ during OAE2 still remains discussed, it has been interpreted as reflecting the
276 input of more radiogenic tethyan seawater at Demerara Rise. As Demerara Rise represented a
277 bathymetric high (Loncke *et al.*, 2020), it may have been potentially more affected by changes
278 in oceanic currents than other parts of the margin basins. Unfortunately, no seawater ϵ_{Nd} data
279 are available at Demerara Rise for the period encompassing the three clay samples that
280 present a slight negative Ce anomaly, a slight HREE enrichment, and more radiogenic $\epsilon_{Nd(t)}$
281 values. Thus, it is not possible to determine if this interval was associated with more
282 radiogenic seawater $\epsilon_{Nd(t)}$ values at Demerara Rise as it was observed for OAE2 and the late
283 Maastrichtian, that could explain a deviation of clay ϵ_{Nd} toward more radiogenic values if
284 authigenic clays formed at that time and integrated this more radiogenic seawater signal.
285 Alternatively, post-depositional alteration of volcanogenic material linked with active
286 volcanism from the CLIP (Caribbean Large Igneous Province) and/or the Bahamas hot spot
287 (Roddaz *et al.*, 2021) would also result in authigenic smectite formation with a quite
288 radiogenic Nd signature. Sporadic excursions of clay Nd isotope composition toward more
289 radiogenic values have also been reported by Roddaz *et al.* (2021), interpreted as reflecting
290 either an increase in the proportion of radiogenic material eroded from ancient mafic sources

291 exposed on the continent, or as the occurrence of volcanogenic inputs. Such input of
292 mafic/volcanogenic material would be expected to induce an Eu anomaly in the REE spectra
293 of the 3 samples with more radiogenic $\epsilon_{Nd(t)}$ values, which is not observed here (Fig. 4;
294 supplementary figure 3).

295 According to the literature, the sediment interval associated with the most radiogenic
296 sample (Core 18 section 7 at 494.26 m ; Fig. 3) also contains a glauconitic level interpreted as
297 an evidence of very low sedimentation rates (Friedrich & Erbacher, 2006 ; Nederbragt *et al.*,
298 2007 ; März *et al.*, 2009). This condensed interval is also characterized by the hiatus observed
299 between the Coniacian and the Campanian, which marks the lithological transition between
300 claystones and marly-chalks (Fig. 3). This particular event may have been induced by the
301 Santonian-Campanian transgression that led to more open oceanic conditions (Haq, 2014).
302 The observation of smear slides, performed on two samples from the bottom of the core,
303 including the sample presenting the highest (most radiogenic) Nd values, clearly reveals the
304 occurrence of glauconite (Fig. 7.A), hence confirming localized neofomed clay minerals.
305 Low sedimentation rates and a decrease of detrital supply to the site would have favoured the
306 formation of authigenic smectite and glauconite, potentially enhanced by in situ weathering of
307 volcanogenic material deposited during this period.

308 The presence of glauconite observed in smear slide and previously described in the core
309 (Erbacher *et al.*, 2004), could possibly lead to an overestimation of illite contents as these two
310 clay minerals have overlapping diffraction peaks that cannot be distinguished by XRD
311 analyses. However, smear slide observations show that only some samples of core 18 are
312 marked by the presence of glauconite while the other levels displaying high illite contents
313 (Fig. 3) do not contain glauconite, confirming that most of clays are detrital for the Demerara
314 Rise site. Palygorskite in marine sediments can also be formed during marine authigenic

315 processes, but TEM observations indicate short and broken palygorskite fibres characteristic
316 of a detrital origin (Fig. 7.B).

317 Apart from the 3 samples with some evidence of clay authigenesis and partial
318 incorporation of a seawater signal, all other clay-size fractions present flat shale-normalized
319 rare earth element (REE) patterns and an absence of any Ce anomaly (Ce/Ce^* above 0.7 ;
320 Roddaz *et al.*, 2021 ; Fig. 4, 5) and corresponds to the “Shale” fields in figure 5.B , thereby
321 indicating a detrital origin for the late Cretaceous clays at Demerara Rise, as already pointed
322 out by Roddaz *et al.* (2021). This is also coherent with the interpretation of März *et al.* (2009),
323 who showed, based on major and trace element composition, that detrital sediments deposited
324 at the Demerara Rise during the late Cretaceous were essentially derived from the erosion of
325 the adjacent Guiana-Suriname margin. Overall, clay minerals at Site 1259 appear mostly
326 inherited from nearby continental landmasses, with only limited and sporadic influence of
327 authigenesis (i.e. glauconite, volcanogenic smectite) along the studied sediment sequence.

328 In the bulk sediment, a net increase in quartz abundance also appears in the Santonian-
329 Campanian interval. However, this increase is also matched by higher opal-CT contents (up to
330 58% of the bulk sediment, Fig. 2). Quartz may also have a marine biogenic origin by
331 transformation of opal-CT during early diagenesis (associated to temperature lower than
332 60°C) in marine sequence around 400 meters burial depth (Hesse, 1988). Thus, its
333 concomitant evolution with opal-CT suggests that the increase of quartz is likely associated to
334 biogenic processes and may in this case correspond to microquartz associated to opal-CT both
335 deriving from opal-A (Riech & von Rad, 1979; Hesse, 1988). It has also been reported that
336 calcareous sediments favour the direct transformation of opal-A into quartz while
337 transformation of opal-CT into quartz is enhanced if detrital quartz is present (Hesse, 1988),
338 which may explain the net increase of quartz in the Santonien-Campanien. For this interval,
339 sediment is probably composed of mixed detrital and biogenic microquartz, impeding the use

340 of quartz content to track variations in detrital inputs. Zeolites (i.e. clinoptilolite, Fig. 2) and
341 opal-CT are also formed authigenically by dissolution of amorphous SiO₂ most likely derived
342 from dissolution-reprecipitation of siliceous organisms (März et al., 2009). We have therefore
343 calculated the ratio (Opal+Clinoptilolite)/Calcite ((O+C)/Ca ; Fig. 8) in order to track the
344 variability of marine primary productivity, as it was demonstrated that clinoptilolite was a
345 good proxy of enhanced biogenic silica productivity (e.g. Karpoff et al., 2007).

346

347 5.2. Sources and detrital material provenance

348 The fine-grained detrital material that is eroded from the exposed continental surface and
349 subsequently transported by rivers is assumed to provide a Nd isotopic composition
350 representative of the corresponding catchment areas (Goldstein *et al.*, 2003; Bayon *et al.*,
351 2015). In our dataset, apart from 3 samples showing some evidence of authigenic clays that
352 will not be considered further to discuss the evolution of the chemical weathering, all other
353 studied clay-size fractions at Site 1259 present geochemical and petrographical features
354 typical of a detrital origin. Isotopic geochemistry of these sediments can therefore be used to
355 track the provenance of the upper Cretaceous sediments deposited on Demerara Rise and to
356 reconstruct the evolution of paleoenvironments in corresponding source regions.

357 To constrain sediment provenance, we use the regional ϵ_{Nd} compilation of detrital sources
358 published by Roddaz *et al.* (2021), applying to their published $\epsilon_{Nd(0)}$ values the same age
359 correction than to our own samples at 83 Ma, in order to compare them to our dataset (Fig.
360 1.B).

361 The units forming the Guiana Craton display the most unradiogenic values (Fig. 1.B).
362 Three units can be distinguished: 1) the Archean Amapa & Imataca granitoids (AAIG), with
363 an average $\epsilon_{Nd(t)}$ value of -30.4 (from -41.5 to -1.5; $2\sigma = 17.2$); 2) the Paleoproterozoic
364 granitoids, gneiss and felsic volcanism (PGGF), with an average $\epsilon_{Nd(t)}$ value of -22.2 (from -

365 41.5 to 30.5; $2\sigma = 17.2$); and 3) the Paleoproterozoic high-grade granulite belts (PHGB), with
366 an average $\epsilon_{Nd(t)}$ value of -19.8 (from -35.4 to -1.8; $2\sigma = 16.2$). The overlying Mesoproterozoic
367 granites (MG) present an average $\epsilon_{Nd(t)}$ value of -17.3 (from -20.0 to -14.5; $2\sigma = 3.9$). Finally,
368 three different sources are characterized by more radiogenic values: the Greenstone Belt (GB)
369 and the Paleoproterozoic mafic intrusions (PMI) yielding values around -7.1 (from -17.6 to
370 4.3; $2\sigma = 13.2$) and the Jurassic mafic dykes (JMD) presenting values around 1.0 (from -4.1 to
371 -6.0; $2\sigma = 5.9$).

372 Our detrital clay $\epsilon_{Nd(t)}$, which range between -17.5 and -14 (Figs. 3, 8) with a mean value of
373 -16.08 ± 0.91 (1σ), do not display any particular trend downcore (Table 2). They also agree
374 well with those reported by Roddaz *et al.* (2021) for a more proximal site, between about -20
375 and -13.5 (Fig. 8). The peripheral regions of the Guiana Shield bordering the Atlantic margin
376 are mainly composed of Paleoproterozoic granitoids and gneisses (PGGF; Fig. 1.B) that
377 represented the main source of the detrital material brought to Site 1259 during the late
378 Cretaceous, with a potential additional contribution of more radiogenic greenstone belts (GB)
379 (Roddaz *et al.*, 2021).

380 The slightly more radiogenic composition of clays during the Turonian could reflect a
381 larger contribution of material derived from the greenstone belts (-7.1; Fig.1.B) or from
382 Jurassic mafic dykes (1.0 ; Fig. 1.B), which would have decreased from the Turonian to the
383 Campanian. Apart from the likely decrease in the contribution of relatively radiogenic rocks
384 to the detrital fraction deposited at Site 1259 from the Turonian to the Coniacian, and a slight
385 increase in the Maastrichtian, no major change in the source of sediments is thus depicted in
386 the Nd isotope signal.

387 The detrital fraction of the studied sediments thus mainly arises from the weathering and
388 erosion of the Guiana craton. To some extent, the evolution in relative proportions of primary
389 clay minerals (illite, chlorite) versus secondary clay minerals (smectite, kaolinite) can provide

390 information on tectonic uplift patterns along continental margins, as newly forming
391 topographic reliefs naturally increase proportions of primary mineral assemblages. Indeed
392 illite and chlorite are primary clay minerals resulting from the erosion of igneous,
393 metamorphic and old sedimentary rocks showing limited degree of chemical weathering
394 intensity. They typically form under relatively dry climate conditions or in active tectonic
395 settings characterized by limited chemical weathering processes (Chamley, 1989). Under
396 more humid climate, (semi-arid or alternation of wet and dry seasons) enhanced chemical
397 weathering of silicate minerals typically leads to smectite formation in soils (bisialitisation ;
398 Chamley, 1989 ; Ruffell *et al.*, 2002; Deconinck *et al.*, 2019). Sub-arid or seasonally arid
399 conditions can also favour the formation of palygorskite that precipitates in evaporative
400 basins, such as inland seas, lakes or other peri-marine environments (Callen, 1984 ; Chamley,
401 1989 ; Daoudi 2004). Finally, kaolinite is commonly associated to hot and wet climate,
402 intense chemical weathering and optimal drainage conditions (Chamley, 1989; Thiry, 2000;
403 Ruffell *et al.*, 2002; Dera *et al.*, 2009).

404 In northeastern South America, warm and humid conditions most likely prevailed during
405 the late Cretaceous, as inferred from combined field data and climate model investigations
406 (Théveniaut *et al.*, 2002 ; Hay *et al.*, 2012), coherent with the observed dominance of pedoge-
407 nic smectite at the Demerara Rise. The fact that only limited amounts of kaolinite were
408 detected during the Maastrichtian at this equatorial site raises questions. It is possible that
409 climate was not sufficiently humid throughout the year to provide either monosialitisation or
410 allitisation (e.g. bauxite formation) leading to abundant kaolinite-rich deposits. However, this
411 latter hypothesis appears in contradiction with evidence of an extensive lateritization phase on
412 the Guiana Shield during the late Cretaceous (Théveniaut *et al.*, 2002). Alternatively, the
413 absence of kaolinite at the studied site could reflect a differential settling effect in seawater,
414 because kaolinite grains are coarser than smectite particles (Godet *et al.*, 2008). While

415 kaolinite was probably formed onshore in the lateritic soils encountered on the Guiana Shield,
416 its net export to the nearby continental margins could have been hampered by preferential
417 deposition in the flooded shelf area (März *et al.*, 2009). Conversely, due to their smaller size,
418 smectite was likely formed preferentially in flatter continental areas and would have been
419 transported to more distal marine environments. Such a gradient in smectite vs kaolinite
420 proportions due to differential settling from the continental plateau to the oceanic basins is
421 commonly observed in modern environments (Chamley, 1989; Gingele *et al.*, 1998).

422 Similarly, primary coarser clay minerals such as illite and chlorite were most likely
423 preferentially deposited in proximal flooded shelf areas and in the GS and FOZ basins rather
424 than on the topographic high of the Demerara Plateau (Fig. 1.B), that could explain the low
425 proportions of primary clay minerals at the site. This would be coherent with geochemical
426 data and sedimentary fluxes reported by März *et al.* (2009) and Loparev *et al.* (2021),
427 indicating a sediment starvation of the Demerara Plateau at this time and preferential deposi-
428 tion of coarse sediments in the flooded shelf area and in the GS and FOZ basins. Such a
429 differential setting effect, resulting in the preferential transport via surface currents of small-
430 size smectite particles to Demerara Rise site, could explain the large dominance of smectite in
431 studied clay fractions (85 to 100% ; Fig. 2).

432 Due to the very high proportions of smectite relative to other clay minerals, the variation in
433 abundance of clay minerals must be interpreted with caution. The co-evolution of illite and
434 palygorskite contents during the Turonian may indicate reworking of clay minerals derived
435 from old sedimentary and igneous rocks or, alternatively, may be linked to enhanced eolian
436 activity, as it is known that palygorskite can be easily transported by winds in sub-arid
437 evaporitic environments (Pletsch *et al.*, 1996 ; Knidiri *et al.*, 2014). In this case, the higher
438 palygorskite proportions recorded at Demerara Rise could indicate the existence of more arid
439 episodes during the Turonian. Alternatively, palygorskite and illite could also come from the

440 erosion of old Mesozoic sediments derived from sedimentary basins bordering the Guiana
441 Craton, including the Takutu Basin (Fig. 1B). Indeed, palygorskite is commonly associated
442 with the Jurassic and early Cretaceous evaporitic lacustrine environments that formed during
443 the opening of the Central Atlantic (Crawford *et al.*, 1984 ; Castro *et al.*, 2021). The erosion
444 of such Mesozoic basins during the Turonian would be consistent with the pre-Coniacian
445 catchment areas defined by Roddaz *et al.* (2021) based on ϵ_{Nd} and trace element abundances.
446 In any case, these minerals represent a very low proportion of the total clay content that is
447 mainly dominated by detrital contemporaneous smectites and should not impact Nd-Hf
448 isotopic values.

449 During the Turonian, the higher content of palygorskite and illite, if eroded from old
450 Mesozoic sediments, could be indicative of higher erosion rates. A slight increase in illite
451 proportions is also recorded during the Coniacian-Campanian interval, with a maximum value
452 around 80 Ma ago, coupled with slightly higher feldspar contents in the bulk sediment (Fig.
453 2). This is also illustrated by the variations of the (I+C)/S and Feld/Clays ratio in figure 8.
454 Although discrete, these features may reflect enhanced mechanical erosion promoting both
455 primary silicate minerals (e.g. feldspar ; Figs. 2, 8) and coarse and primary clay minerals (e.g.
456 illite) during an uplift phase. This would be consistent with the timing of the main pulses of
457 tectonic activity and higher sedimentation rates in the GS and FOZ basins during this interval
458 (Sapin *et al.*, 2016 ; Loparev *et al.*, 2020 ; Fig. 1.B). Presumably, at that time, enhanced
459 tectonic activity along the northeastern South American margin was accompanied by
460 enhanced erosional fluxes and nutrient export to seawater, which could also have driven
461 enhanced biosiliceous marine productivity inferred from the (O+C)/Ca ratio (Fig. 8). This
462 mechanism may explain the concomitant increase of biogenic phases and the detrital input
463 during the Campanian in relation to the main tectonic pulse, although a role of aerial

464 explosive volcanism activity as a potential source of siliceous material cannot be excluded
465 (Frogner *et al.*, 2001 ; Stokke *et al.*, 2021).

466

467 5.3. Evolution of chemical weathering linked to climate and tectonic.

468 The identification of distinct uplift pulses during the Turonian and Campanian-to-early
469 Maastrichtian period in the studied area (Fig. 9, Sapin *et al.*, 2016) has provided evidence for
470 a major tectonic phase affecting the eastern South American margin at that time. Argon/argon
471 (Ar/Ar) dating of the late Cretaceous planation surface S0 (McConnell, 1968 ; Sapin *et al.*,
472 2016) yields an age of 71 Ma, while apatite fission track data (AFTA) indicate ages of 77 ± 3
473 Ma, 83 ± 5 Ma and 99 ± 9 Ma for the periods of enhanced regional tectonic activity (Fig. 9 ;
474 Sapin *et al.*, 2016). In the FOZ basin, the highest terrigenous accumulation rates occurred
475 during the Campanian-Maastrichtian interval (Loparev *et al.*, 2021), associated with the main
476 uplift phase, while in the GS basin, Roddaz *et al.* (2021) shows an increase in sedimentation
477 rates during the Santonian.

478 In order to track the impact of tectonic uplift, inferred from Ar/Ar dating of planation
479 surfaces and AFTA, on the evolution of chemical weathering intensity in NE South America,
480 we now consider Nd and Hf isotope data measured in the clay-size fraction. The combined
481 use of Nd and Hf isotopes in fine-grained sediment records is a powerful proxy to track
482 changes in continental chemical weathering (Bayon *et al.*, 2012 ; Fontanier *et al.*, 2018 ;
483 Corentin *et al.*, 2022; Bayon *et al.*, 2022). While the Lu-Hf and Sm-Nd isotopic systems
484 behave similarly during magmatic processes, defining a so-called «Terrestrial array» in most
485 igneous and sedimentary rocks (Fig. 6; Vervoort *et al.*, 1999 ; Vervoort *et al.*, 2011), they are
486 decoupled during Earth surface processes such as chemical weathering and sediment transport
487 (Vervoort *et al.*, 1999 ; Bayon *et al.*, 2016 ; Garçon *et al.*, 2013 ; Garçon & Chauvel, 2014 ;
488 Chauvel *et al.*, 2014). This decoupling is partly due to mineral sorting, which causes

489 preferential enrichment of zircons in the coarse-grained sediment fraction, characterized by
490 high Hf concentrations and very low ϵ_{Hf} (unradiogenic) compositions (Vervoort *et al.*, 1999 ;
491 Chauvel *et al.*, 2014). By contrast, clay-size fractions of sorted sediments are typically
492 depleted in zircons and hence are systematically shifted towards more radiogenic ϵ_{Hf} values
493 compared to coarser sediment fractions, defining a so-called « clay array » (Zhao *et al.*, 2014;
494 Bayon *et al.*, 2016 ; Fig. 6). Additionally, the mineral phases most susceptible to chemical
495 weathering on continents are typically enriched in Lu relative to Hf, hence leading to the
496 preferential release of radiogenic Hf during weathering (Bayon *et al.*, 2006 ; Bayon *et al.*,
497 2016 ; Dausmann *et al.*, 2019). Upon formation of secondary clay minerals in soils, a fraction
498 of dissolved radiogenic Hf can be incorporated into clays (Bayon *et al.*, 2016). As a result,
499 clay-size fractions derived from intensively weathered soils are associated with more
500 radiogenic clay ϵ_{Hf} signatures, which partly reflect an increase in the relative proportion of
501 secondary clay minerals (high ϵ_{Hf}) relative to unweathered primary detrital mineral (low ϵ_{Hf}).
502 In fine-grained sediment records, a more intense chemical silicate weathering hence results in
503 a vertical departure of clay ϵ_{Hf} from the « clay array », noted $\Delta\epsilon_{\text{Hf}}$ (Fig. 6 ; Bayon *et al.*,
504 2016). Note that $\Delta\epsilon_{\text{Hf}}$ has not been calculated for the 3 samples presenting evidence of a
505 seawater imprint on the clay fraction Nd isotope signal linked to the presence of authigenic
506 clays.

507 The analysed clay fraction presents low zirconium (Zr) concentrations (between 42 and
508 157 ppm), in the range of modern clay-size fractions separated from river sediments (Bayon *et al.*
509 *et al.*, 2015). Additionally, there is no significant correlation between ϵ_{Hf} values and Zr
510 concentrations ($R^2 = 0.23$; Supplementary Fig. 1), nor between $\Delta\epsilon_{\text{Hf}}$ and Zr concentrations.
511 This observation indicates that the $\Delta\epsilon_{\text{Hf(t)clay}}$ evolution of studied clays is not controlled by the
512 presence of residual zircons in the isolated $< 2\mu\text{m}$ sediment fraction, but rather reflects
513 variations in silicate weathering intensity of the catchment area.

514 The close correspondence of $\Delta\varepsilon_{\text{Hf clay}}$ with climatic parameters and with the chemical index
515 of alteration (CIA) observed in clays recovered from rivers receiving weathering products
516 from source rocks of various nature suggests a minor influence of lithology on $\Delta\varepsilon_{\text{Hf}}$ (Bayon *et*
517 *al.*, 2016). At Site 1259, the limited $\varepsilon_{\text{Nd}(t)}$ variations through the studied interval points to the
518 absence of major change in the lithologies of the sediment source, while $\Delta\varepsilon_{\text{Hf}(t)\text{clay}}$ increases
519 quite markedly. Most importantly, no correlation is depicted between $\varepsilon_{\text{Nd}(t)}$ and $\Delta\varepsilon_{\text{Hf}(t)}$,
520 demonstrating that variations in $\Delta\varepsilon_{\text{Hf}(t)}$ are not controlled by lithology variations of the source
521 rock at this site (Supplementary figure 2).

522 Calculated $\Delta\varepsilon_{\text{Hf}(t)\text{clay}}$ values at Demerara Rise are thus interpreted hereafter to reflect the
523 evolution of chemical weathering intensity, with the depicted decrease in $\Delta\varepsilon_{\text{Hf}(t)\text{clay}}$ pointing
524 to a decrease in weathering of surrounding catchment regions from the end of the Turonian to
525 the Santonian or beginning of Campanian (Fig. 8 ; Bayon *et al.*, 2016). Then, from
526 Campanian to the end of Maastrichtian, a marked increase in regional silicate weathering
527 intensity is recorded (Figs. 3, 8). Superimposed on this trend, a slight decrease in $\Delta\varepsilon_{\text{Hf}(t)\text{clay}}$
528 values, reflecting an interval of lower weathering intensity, is also recorded during the late
529 Campanian and Early Maastrichtian, of about 2.0 (Figs. 3, 8).

530 The Chemical Index of Alteration (CIA) has also been widely used to track changes in
531 chemical weathering (e.g. Young & Nesbitt, 1999; Roddaz *et al.*, 2006), as it provides an
532 estimation of the degree of mobile element depletion relative to immobile elements during
533 chemical weathering (Nesbitt & Young, 1982), although this proxy is known to be influenced
534 by sediment source and mineral sorting (Fantasia *et al.*, 2019). At Site 1259, no major change
535 in sediment provenance occurred during the late Cretaceous but Campanian sediments are
536 marked by higher opal-CT and quartz contents increasing the concentrations of silicon and
537 thus decreasing aluminium relative proportions. The CIA value calculated for the two samples
538 exhibiting the highest contents of biogenic minerals is very low (<50), hence suggesting a

539 dilution effect by Si impeding its use as a weathering proxy in the interval marked by high
540 opal-CT and quartz contents (Fig. 8). The pronounced increase of the CIA that subsequently
541 follows (from 56 to 74) is however not associated to concomitant change in opal-CT and
542 quartz contents, nor in sediment sources as ϵ_{Nd} values remain quite constant, and thus could
543 reflect an increase of chemical weathering intensity from the middle Campanian to the late
544 Maastrichtian. This interpretation would be in agreement with the increase depicted in
545 $\Delta\epsilon_{Hf(t)clay}$.

546 Over long timescales, the evolution of continental chemical weathering is thought to be
547 driven mostly by climate and tectonics. Increasing temperature and precipitation favour the
548 kinetics of silicate weathering reactions and increase wetting of reactive mineral surface areas,
549 enhancing silicate mineral dissolution rates (White & Blum, 1995 ; Berner *et al.*, 1983 ;
550 Gaillardet *et al.*, 1999 ; Barron & Washington, 1985 ; Pierrehumbert, 2002). Active tectonic
551 settings associated with high denudation rates and mechanical erosion can also promote high
552 chemical weathering rates by favouring both the disaggregation of silicate minerals and
553 renewing the exposure of fresh rock surfaces available to weathering. The relationship
554 between mechanical erosion and chemical weathering is however not straightforward. Under
555 supply-limited weathering conditions, the rate of chemical weathering is proportionally
556 related to denudation rates (Raymo *et al.*, 1988 ; West *et al.*, 2005 ; Gabet & Mudd, 2009). In
557 contrast, under kinetically-limited weathering conditions, detrital material is removed too fast
558 from the regolith, and higher denudation rates are not accompanied by higher chemical
559 weathering rates (West, 2005 ; Gabet and Mudd, 2009).

560 The evolution of chemical weathering intensity depicted by $\Delta\epsilon_{Hf(t)clay}$ matches relatively
561 well the evolution of ocean temperature inferred from both planktic foraminifera at Demerara
562 Rise (Fig. 8 ; Bornemann *et al.*, 2008) and the global benthic foraminiferal $\delta^{18}O$
563 reconstruction (Fig. 8 ; Friedrich *et al.*, 2012). Chemical weathering in northeastern South

564 America markedly decreased during the climate cooling recorded from the end of the
565 Turonian to the early Campanian, suggesting a climatic control of this evolution, as lower
566 temperature reduces silicate weathering reactions (Fig. 8 ; White and Blum, 1995). Similarly,
567 the cooling recorded in the latest Campanian and early Maastrichtian appears to be also
568 associated with a decrease, albeit more limited, of continental weathering intensity (Fig. 8).
569 However, if a slight warming is observed in the foraminiferal $\delta^{18}\text{O}$ dataset during the middle
570 and late Campanian, global temperature remains markedly lower in the late Campanian and
571 Maastrichtian than during the Turonian, when chemical weathering markedly increased as
572 inferred from $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ values exceeding those recorded for the Turonian. Therefore, for the
573 middle Campanian to Maastrichtian interval, the evolution of global temperature is unlikely to
574 fully account for our chemical weathering record, hence requiring an additional process
575 resulting in enhanced silicate weathering. Throughout the studied interval, there is no
576 evidence for marked changes in the amount of precipitation; nearby continental regions
577 remained within a humid climatic belt as documented by the occurrence of laterites and
578 bauxites throughout the late Cretaceous (Théveniaut *et al.*, 2002 ; Hay, 2012). Consequently,
579 we infer that the observed marked increase in chemical weathering was presumably not
580 induced by a regional climate shift towards wetter conditions.

581 As mentioned above, during the Campanian, the northeastern South American margin
582 experienced several tectonic pulses that led to the uplift of the craton (McConnell, 1968 ;
583 Potter, 1997 ; Harman *et al.* 1998 ; Morais Neto *et al.* 2009 ; Peulvast and Bétard 2015 ; Sapin
584 *et al.*, 2016 ; Monteiro *et al.* 2018). This uplift most likely drove enhanced mechanical erosion
585 as suggested by the slight increase in illite and feldspar at Site 1259, which, in a permanently
586 humid climatic belt, could have been associated with more intense silicate weathering as well
587 (West, 2005 ; Gabet & Mudd, 2009). The occurrence of a major uplift phase concomitant with
588 a pronounced $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ increase (Fig. 8), with values exceeding those recorded in the Turonian

589 despite markedly lower temperatures, strongly points towards tectonics as being the main
590 driver of chemical weathering variability in northeastern South America during the middle
591 Campanian to Maastrichtian interval. By contrast, from the end of the Turonian to the early
592 Campanian interval, when tectonic activity was reduced, climate was instead the main
593 parameter controlling the evolution of chemical weathering in the study area.

594 Altogether, our data thus suggest the dominant control of tectonics on continental
595 weathering when occurring under wet climate conditions, with a more limited role of
596 temperature. These findings complement previous inferences based on Site 356 offshore
597 Brazil, which showed that under semi-arid climate conditions, chemical weathering did not
598 respond to the tectonic uplift of the Brazilian margin until local climate evolved toward more
599 hydrolysing conditions (Corentin *et al.*, 2022). Surprisingly, the $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ values recorded at
600 site 1259 are negative for the most part, with values down to -5 which are observed at present
601 in cold and dry climatic zones, although values recorded during the latest Campanian and
602 Maastrichtian, up to 1, are observed in modern tropical humid belts (Bayon *et al.* (2016). By
603 contrast, higher $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ values are recorded at site 356 located in a semi-arid climatic zone
604 (Fig. 6). This apparent discrepancy may further point to a preponderant role of tectonics on
605 silicate weathering through supply of fresh minerals exposed to weathering processes,
606 favoured by enhanced physical erosion. At site 1259, in the context of relative tectonic
607 quiescence during the interval recording the most negative values, these low $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ values
608 may reflect the existence of limited chemical weathering under supply-limited conditions
609 (West *et al.*, 2005; Gabet and Mudd, 2009), impeded by low physical erosion rates reducing
610 the renewed supply of fresh minerals to weathering processes. Alternatively, the
611 correspondence between absolute $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ values with climatic zones may have been
612 different in the overall greenhouse climate mode of the late Cretaceous than from our modern
613 icehouse climate mode, in which they have been established. Although the significance of

614 absolute $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ values still needs further investigation, our study highlights the potential of
615 relative variations in $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ at a specific site remains to infer relative changes in silicate
616 chemical weathering at a local scale.

617 Variations in continental weathering can also impact global climate through atmospheric
618 CO_2 drawdown by silicate weathering reactions. Combined Hf-Nd and clay mineralogical
619 data from Site 356 offshore Brazil have recently highlighted an increase in silicate weathering
620 along the Brazilian margin from the Santonian onward, which coincided with an acceleration
621 of the late Cretaceous global climate cooling (Friedrich *et al.*, 2012). At Site 1259, the marked
622 increase in chemical weathering affecting the northeastern South American margin initiated
623 during the middle Campanian, appears post-dating the main global cooling phase inferred
624 from the benthic foraminiferal $\delta^{18}\text{O}$ record. While the tectonic uplift of the northern South
625 American margin and its impact on chemical weathering hence could not have triggered the
626 long-term climate cooling during the late Cretaceous, it is possible that weathering-driven
627 atmospheric CO_2 drawdown contributed to maintaining cooler conditions in the late
628 Campanian and Maastrichtian. Altogether, the data from Site 356 (Corentin *et al.*, 2022)
629 offshore southeastern Brazil, and the new data from Site 1259 at Demerara Rise (this study)
630 highlight the possible importance of the late Cretaceous uplift of the Eastern South American
631 margin on the long-term cooling recorded during this period.

632

633 *Conclusion*

634 The new $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ proxy based on combined Nd-Hf isotope measurements of clay-size
635 fractions was applied to a sediment record from Demerara Rise in order to reconstruct the
636 evolution of continental silicate weathering intensity at the northeastern South American
637 margin during the late Cretaceous.

638 The depicted evolution of $\Delta\epsilon_{\text{Hf}(t)\text{clay}}$ seems to show a co-variation of chemical weathering
639 with global climate, except during the middle Campanian to Maastrichtian interval when
640 inferred chemical weathering intensity was higher than during the Turonian despite cooler
641 climate conditions. During this Campanian-to-Maastrichtian period, our proxy record
642 indicates that major tectonic uplift of the northeastern South American margin was
643 accompanied by an intensification of chemical weathering. By contrast, from the end of the
644 Turonian to the early Campanian, i.e. a period characterized by reduced tectonic activity, the
645 evolution of continental weathering in northern South America was mostly influenced by
646 climate.

647 Our dataset brings new constraints on the combined influence of tectonics and climate in
648 driving the long-term evolution of continental chemical weathering through time. Under
649 persistent tropical climate conditions, tectonics appears to play a major role in controlling
650 silicate weathering, while the impact of temperature appears less prominent. Overall, the
651 response of chemical weathering to tectonic uplift along the eastern margin of South America
652 inferred from our proxy data suggests that this major tectonic event could have partially
653 contributed to the late Cretaceous cooling through weathering-driven atmospheric CO_2
654 drawdown.

655

656 *Data availability*

657 All the data are provided in the supplementary materials attached to this paper.

658

659 *Acknowledgments*

660 We thank IODP for providing access the material. IODP France and ANR RISE funded the
661 project. We also thank Rémi Chassagnon (Laboratory ICB, University of Burgundy) and

662 INRAe of Dijon, Ludovic Bruneau of the Biogéosciences Laboratory and GISMO analytical
663 Platform (University of Burgundy), Marie-Laure Rouget and Bleuenn Gueguen of the Pôle
664 Spectrométrie Océan (Brest) and Philippe Telouk of the ENS Lyon for their analytical
665 support.

666

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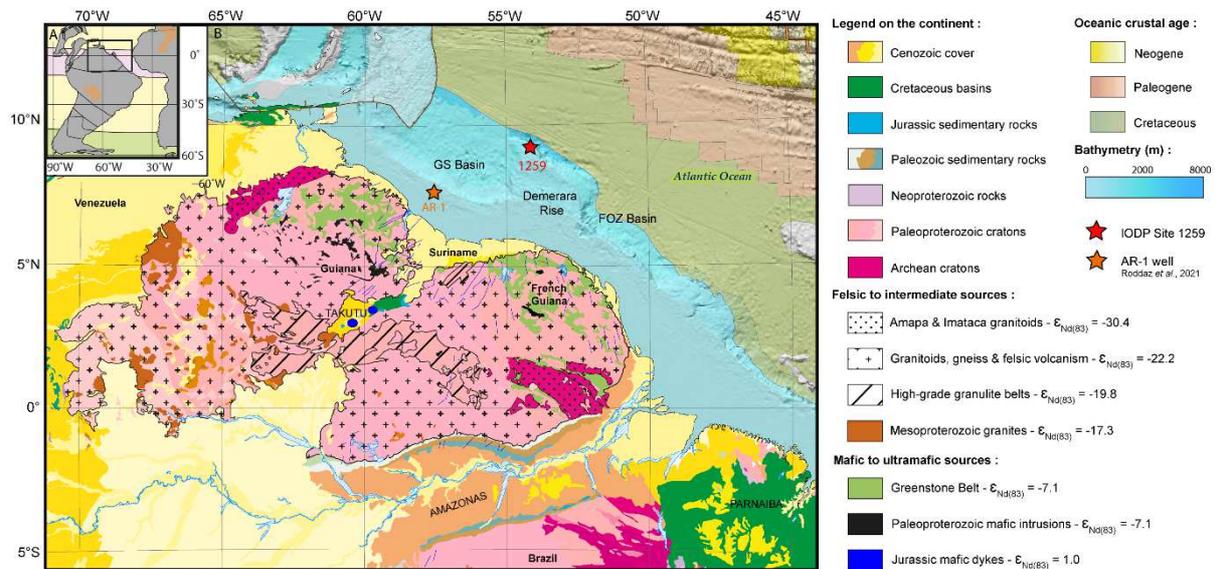
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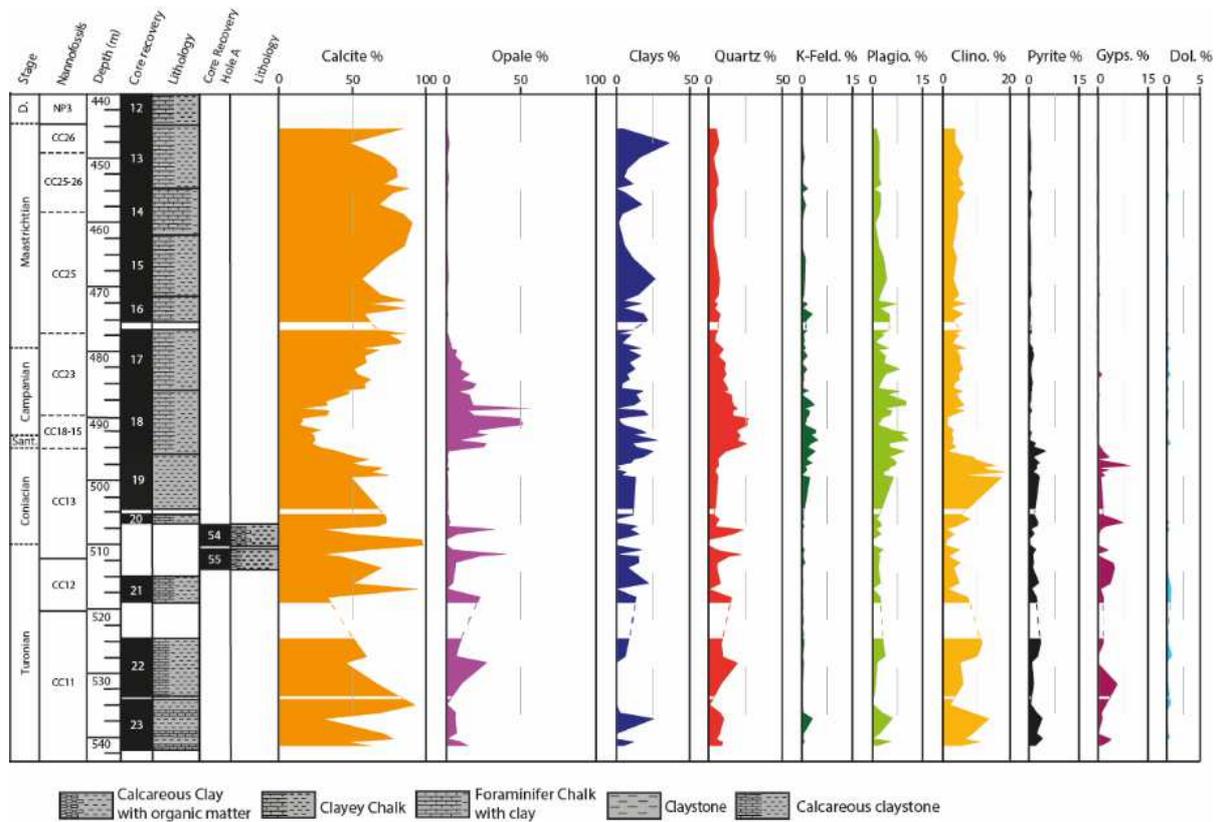
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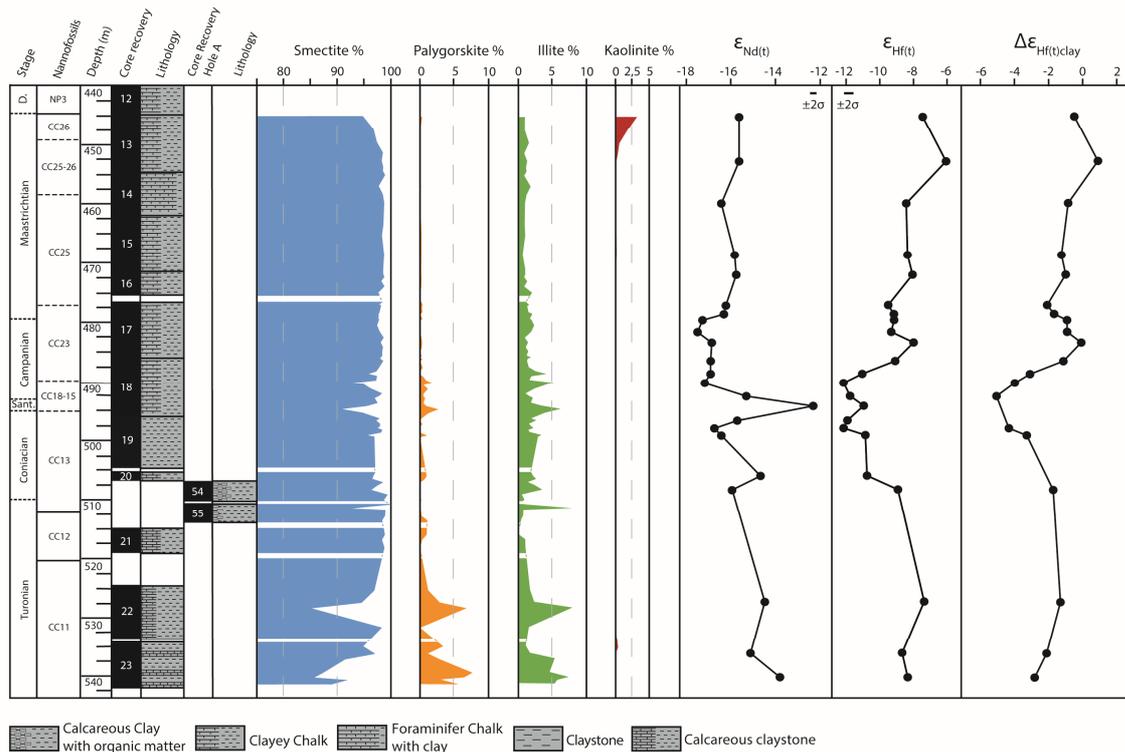


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944 Figure 1 – A. Palaeogeographic map of the South American continent for the Campanian (83
 945 Ma ; www.odsn.de) with the humid climatic belt in pink, the arid climatic belt in yellow, the
 946 mid-latitude warm humid belt in green and the evaporitic belts in orange (according to Hay &
 947 Floegel, 2012). B. Geological map of the northeastern Brazilian margin with the main $\epsilon_{Nd(t)}$

948 units of the Guiana Shield from Roddaz *et al.* (2021) calculated at 83 Ma and location of Site
 949 1259 and AR-1 well from Roddaz *et al.*, 2021.





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955 Figure 3 – Lithology, biostratigraphy, clay mineralogy, $\epsilon_{Nd(t)}$, $\epsilon_{Hf(t)}$ and $\Delta\epsilon_{Hf(t)clay}$ of ODP Site
 956 1259. Abbreviations : Sant. = Santonian, D. =Danian. The black bars corresponds to the
 957 external reproductibility (2 s.d) of ϵ_{Nd} and ϵ_{Hf} based on repeated analyses of JMC 475 and
 958 JNDi standards.

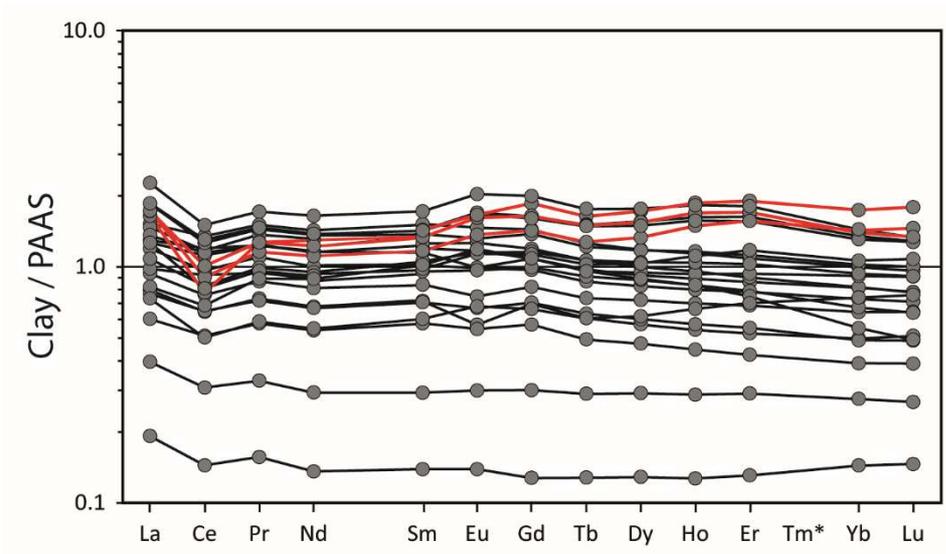
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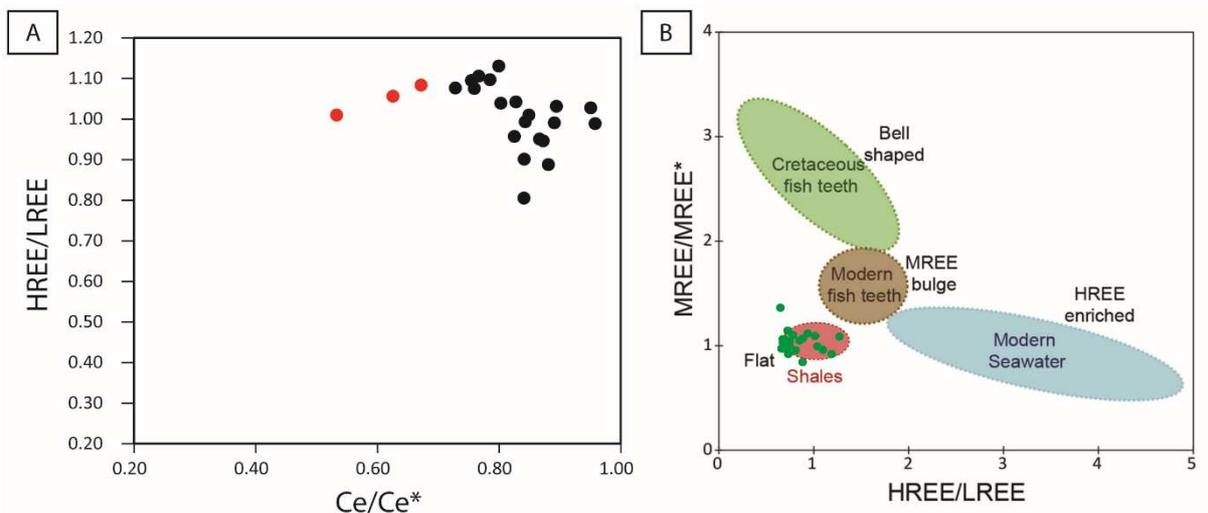
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965 Figure 4 – Shale-normalized (PAAS ; Taylor and McLennan, 1985) REE patterns for the clay
 966 fraction of Site 1259. The three samples in red presents a slight negative Ce anomaly and a
 967 slight enrichment in HREE.

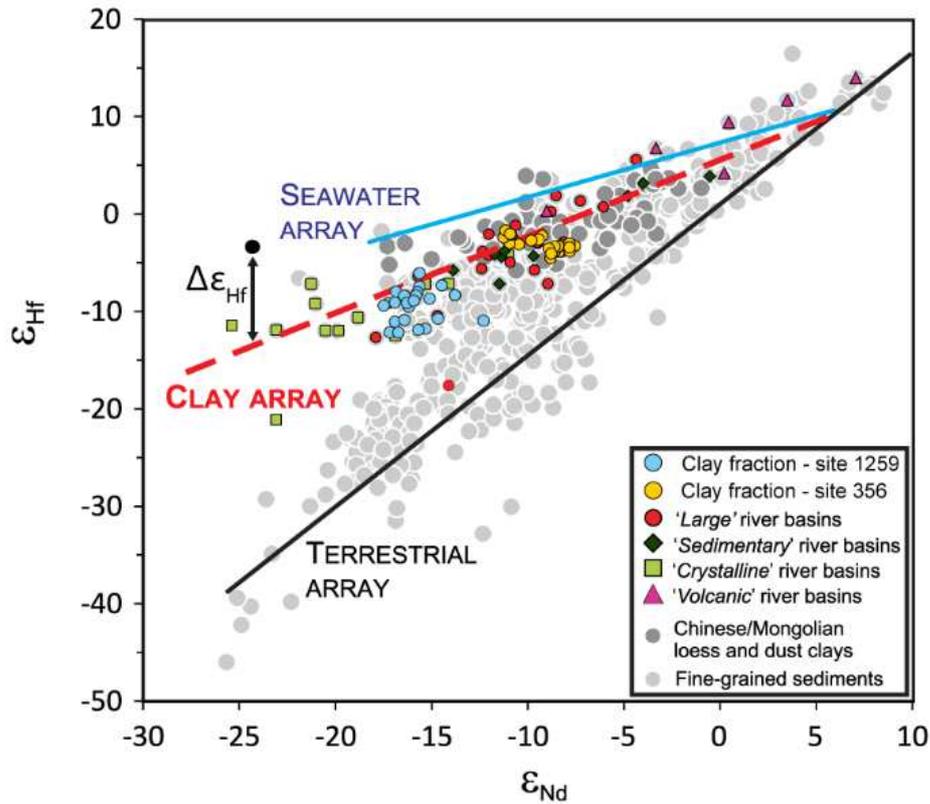
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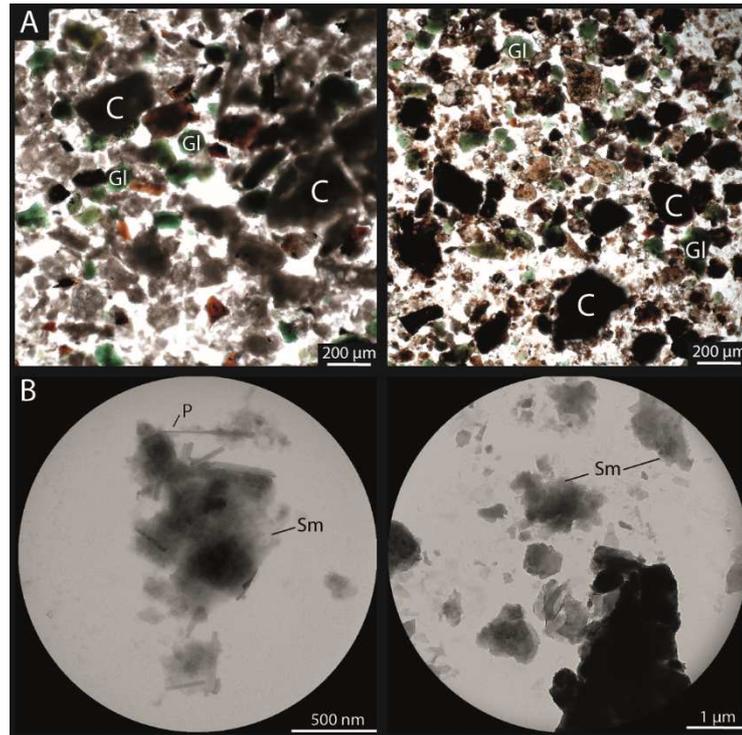
970 Figure 5 – A. HREE/LREE vs Ce/Ce* diagram with the three samples in red presenting the
 971 most negative cerium anomaly. B. MREE/MREE* vs HREE/LREE diagram. The data of this
 972 study appears in green dots and were potted on the diagram of Roddaz et al., 2021. The
 973 samples plotted in the “flat” REE patterns end member characteristic of continental clays.

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 976 Figure 6 – Nd and Hf isotopic composition of clay-size fractions at Site 1259 and Site 356
 977 (Corentin et al., 2022) together with data for modern river clays (modified from Bayon *et al*,
 978 2016). The Clay Array corresponds to the correlation displayed by ϵ_{Nd} and ϵ_{Hf} in modern clay-
 979 size fractions (Bayon *et al.*, 2016). $\Delta\epsilon_{\text{Hf}}$ corresponds to the vertical ϵ_{Hf} deviation of any
 980 sample relative to the clay array, which reflects the intensity of chemical weathering.

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992 Figure 7 – A – Smear slides of the samples 18r7w31 (left) and 18rCCw3 (right) containing

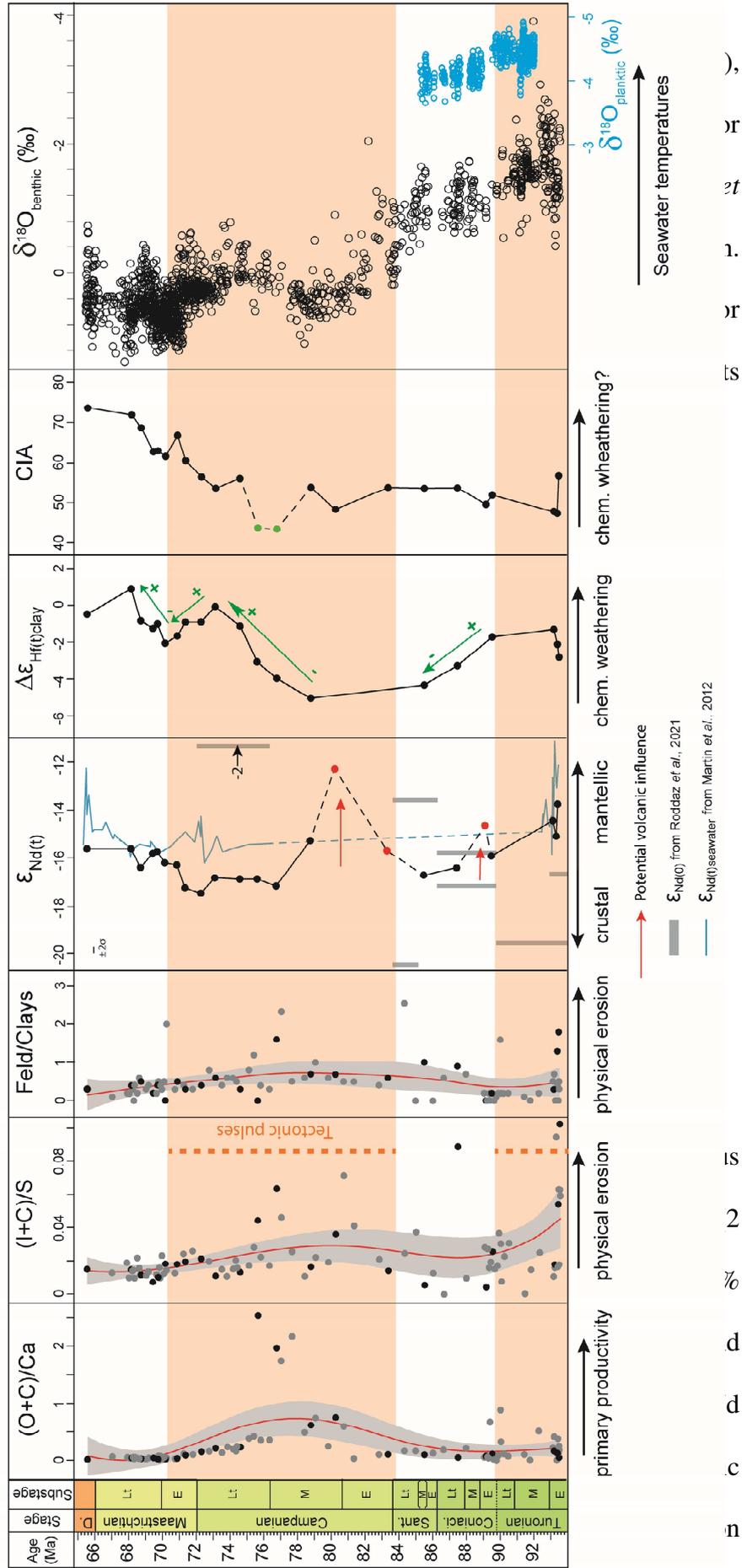
993 clay minerals (dark brown) noted “C” and glauconite aggregate (green) noted “Gl”. B –

994 Transmission electron micrographs (TEM) of detrital particles of smectite with flake shape

995 (left, sample 22r3w100) and short fibres of detrital palygorskite with smectite particles (right,

996 sample 18r7w80).

997 Figure 8 –Evoluti
 998 (Illite+Chlorite)/Smectite
 999 Site 1259 clay-size fractio
 1000 *al.*, 2012) and local $\delta^{18}\text{O}_1$
 1001 The grey and black dots
 1002 which both geochemical a



1003 correspond to literature d
 1004 values (see text for discus
 1005 s.d) of ϵ_{Nd} on repeated
 1006 confidence interval have t
 1007 Feld/Clays ratios, with a l
 1008 isotopic compositon of R
 1009 composition of Martin et a

1010 of chemical weathering on the margin. Abbreviations : Coniac. = Coniacian, Sant. =
1011 Santonian, D. = Danian, E = Early, M = Middle, Lt = Late, chem. = chemical.