

Provenance constraints on the Cretaceous-Paleocene erosional history of the Guiana Shield as determined from the geochemistry of clay-size fraction of sediments from the Arapaima-1 well (Guyana-Suriname basin)

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14 Abstract

Recent studies have emphasized the role of the Equatorial Atlantic Ocean opening on the 15 drainage reorganization and erosion of the West African and northeastern Brazilian cratons 16 during the Cretaceous. However, such studies are lacking regarding the Guiana Shield. To fill 17 this gap, we determined the provenance of sediments deposited in the Suriname-Guyana basin 18 during the Cretaceous-Paleocene based on the total organic carbon (TOC) content, $\delta^{13}C_{TOC}$, 19 major and trace element concentrations, and Sr-Nd isotopic data of nineteen clay-size fraction 20 samples from one industrial well (Arapaima-1). Overall, the absence of correlation between the 21 TOC content, $\delta^{13}C_{TOC}$ and provenance proxies as well as the lack of Ce anomaly indicate that 22 marine authigenesis, reducing conditions, and weathering have not markedly affected the trace 23 element ratios (Eu/Eu*, Cr/Th, Th/Sc) and Sr-Nd isotopic compositions used as provenance 24 proxies. Three samples (Early Cretaceous, Cenomanian and Late Campanian) have radiogenic 25 $\varepsilon_{Nd(0)}$ values >-2.2. The Early Cretaceous sample may have been sourced by Precambrian or 26 Mesozoic mafic rocks of the Guiana Shield or contemporaneous basic volcanic rocks. The 27 Cenomanian sample is likely to have been sourced by basic volcanic rocks of the Caribbean 28 Large Igneous Province or the Takutu rift whereas the Late Campanian sample may have 29 recorded more acid volcanism in the Takutu Rift. The other samples have an overall Guiana 30 31 cratonic provenance with variable contributions of the Paleoproterozoic and Mesoproterozoic terranes and Jurassic or Precambrian mafic dykes. Increases in ⁸⁷Sr/⁸⁶Sr ratios and trace element 32 ratios characteristic of more differentiated sources suggest an increasing contribution from the 33 inner part of the Guiana Shield after the Coniacian. Because of the possible presence of active 34 volcanism during the deposition of analyzed samples, post rift mantle upwelling under the 35

36 equatorial margin is the favored mechanism to explain the increasing erosion of the Guiana

- 37 Shield inner parts.
- 38 Keywords: Provenance, Cretaceous, Guiana Shield, Guyana-Suriname Basin,

40 1. Introduction

41 The structure and geodynamic evolution of transform continental margins have recently received a lot of attention (i.e. Basile, 2015; Mercier de Lépinay et al., 2016; Nemčok et al., 42 2016). In particular, the conjugate West African, Suriname, Guyana, French Guiana and 43 Brazilian equatorial margins are among the best examples (Basile et al., 2005; Emery et al., 44 1975; Mascle and Blarez, 1987; Mercier de Lépinay et al., 2016). Whereas the kinematics 45 46 evolution of the Equatorial Atlantic Ocean and its margins is well constrained in time and space (Heine et al., 2013; Moulin et al., 2010), less attention has been paid to the erosional history of 47 adjacent source areas (i.e., West African and Guiana and Brazilian cratons or Phanerozoic 48 49 basins). Noticeable exceptions regarding the African margin is the provenance study of Mourlot et al. (2018a) which emphasized that the West African Craton (WAC) may be viewed as a 50 dynamic area during the Cretaceous opening of the Equatorial Atlantic Ocean. Using the Nd 51 isotopic composition of marine sedimentary rocks sampled offshore along the African margin, 52 these authors have highlighted that the erosion the WAC evolved from its outer parts in the 53 Albian–Middle Cenomanian interval to its inner parts in the Campanian–Maastrichtian (Mourlot 54 et al., 2018a). 55

Like other Atlantic passive margins, the Brazilian and Guyana-Suriname margins are bordered by elevated low relief inland plateaus separated from a a higher relief coastal plain by one or more steep escarpments (Cogné et al., 2012; Gallagher et al., 1994; Gallagher and Brown, 1999; Hiruma et al., 2010; Japsen et al., 2012; Jelinek et al., 2014; Tello Saenz et al., 2003). The presence of a Late Cretaceous "peneplanation surface" in the inner part of the Guiana Shield cropping out at elevation > 900 m asl is ascribed to Campanian erosion that may have affected this part of the Guiana Shield (Sapin et al., 2016). In northeastern Brazil, various

thermochronological and geomorphological studies have presented evidences of Late Cretaceous
uplift (Harman et al., 1998; Morais Neto et al., 2009; Peulvast and Bétard, 2015; Potter, 1997).
However, all these studies are based on mapping or sampling of isolated Cretaceous outcrops
and this may question the representativeness of these studies at basin scale. To date there is no
provenance study dealing with the Cretaceous erosional history of the Guiana Shield.

68 Determining the provenance of well dated deep-sea sediments deposited in marginal basins may be particularly useful to provide constraint on the erosional history of cratons (Clift 69 and Blusztajn, 2005; Mourlot et al., 2018a). Many tools exist to determine the provenance of 70 71 sedimentary rocks including sandstone petrography, clay mineralogy, heavy mineral analyses, geochronological dating on accessory minerals, major and trace element geochemistry and Sr-72 Sm-Nd isotopic compositions of bulk sedimentary rocks (see for instance Basu et al., 1990; 73 Caracciolo, 2020; Cullers, 2000; Dickinson, 1985; Dickinson and Suczek, 1979; Garzanti, 2016, 74 2019; Garzanti and Andò, 2019; McLennan et al., 1993a; Weltje, 2012 and references therein). 75 76 All of these tools are subjected to biases related to source-rock mineral fertility (see for instance Chew et al., 2020; Dickinson, 2008; Malusà et al., 2016, 2013), pre and post depositional 77 processes including weathering, grain-size sorting and diagenesis (see for instance (Bouchez et 78 79 al., 2011; Fedo et al., 1995; Garzanti et al., 2008, 2009, 2014; Lawrence et al., 2011; McLennan et al., 1993a; Moquet et al., 2020; Nesbitt et al., 1996; Roddaz et al., 2006, 2014; Rousseau et al., 80 2019; Viers et al., 2008) and analytical procedures (Andersen et al., 2019; Chew et al., 2020; 81 82 Malusà et al., 2013; Vermeesch, 2018, 2004). In addition, the low amount of material available for well or core sample makes geochronological dating methods (especially U-Pb dating on 83 84 zircon) difficult to use. Trace elements ratios (i.e. Th/Sc, Cr/Th, Eu/Eu*) as well as Sr-Nd 85 isotopic composition of clay size sediments may be preferred to establish a complete integration

of drainage pathways from the most distant cratonic areas to oceanic basins (Patchett et al., 86 1999) because they are not biased by grain-size sorting (e.g. Cullers, 2000; McLennan et al., 87 1993) and require a lower amount of bulk sediment (~3 g, Bayon et al., 2002) These 88 geochemical approaches have been recently used to determine the source of the Late Eocene to 89 Quaternary sediments deposited in the Mozambique Channel (Roddaz et al., 2020) or to 90 reconstruct the Cretaceous erosional history of the WAC (Mourlot et al., 2018). Furthermore, the 91 Nd isotopic composition of past sediments deposited in ocean margins can provide constraints 92 for determining the influence of continental Nd inputs released from river-borne lithogenic 93 94 sediments deposited at ocean margins on Nd isotopic composition of past oceans. Indeed, Rare Earth Elements (REE) release from lithogenic sediments in river discharge may ultimately 95 dominate all other REE inputs to the oceans (Jeandel et al., 2007; Jeandel and Oelkers, 2015; 96 97 Rousseau et al., 2015). Such constraints are crucial for all oceanic circulation reconstruction based on the Nd composition of past oceanic masses (e.g. (Dera et al., 2015; Donnadieu et al., 98 2016; Mourlot et al., 2018b). Currently, the only Nd isotopic data available for the Cretaceous 99 100 sediments of the Guyana and Brazilian margins of the Equatorial Atlantic Ocean are those from 101 Martin et al. (2012). These data were obtained on fish teeth and sediments from the Demerara 102 Rise (ODP Leg 207) which was a bathymetric high during the Cretaceous (e.g. Arthur and Natland, 1979. Loncke et al., 2019) and may have been potentially more influenced by oceanic 103 currents than other parts of the equatorial margin basins. 104

The aim of this study is to determine the provenance of siliciclastic sediments deposited at the Suriname-Guyana basin during the Early Cretaceous-Paleocene based on TOC content, $\delta^{13}C_{TOC}$, major and trace element concentrations and Sr-Nd isotopic data of nineteen clay-size fraction of sediment samples from one industrial well Arapaima-1 (AR-1, Fig. 1). Determining

the provenance of these sediments may help to bridge the gap between provenance studies
regarding West African and Guiana cratons and may provide constraints on the erosional history
of the Guiana craton.

112 **2. Geological setting and sedimentary sources**

113 2.1 Tectonic and geodynamic setting

The Guyana-Suriname basin is located along the passive margin of northeastern South 114 America (Fig. 1). It includes the offshore of Guyana, Suriname and French Guiana. It is 115 116 delimited by the Demerara Plateau to the east, the Barbados Ridge accretionary prism to the 117 west, the Guiana shield to the south and the Demerara Abyssal plain to the north (Fig. 1). The tectonic evolution of the Guyana-Suriname basin can be divided into three main geodynamic 118 periods linked to the opening of the Atlantic Ocean from the Early Jurassic (~200 Ma) to present 119 (Yang and Escalona, 2011). The first geodynamic period is related to the Jurassic rifting between 120 South America and North America. It was first initiated in the Central Atlantic region and 121 122 resulted in east-west extension with a large component of dextral shearing in the Guyana-Suriname basin (Gouyet et al., 1994). This rifting period resulted in the formation of grabens in 123 the present-day offshore region of the Guyana-Suriname basin and in the east-west-oriented 124 Takutu graben separating the Guyana shield in two parts (Fig. 1). The thick (approximately 21 125 km) volcanic basement of the Demerara Rise formed as a result of syn-rift-related volcanism 126 linked to the Bahamas hot spot at the initial opening of the Central Atlantic (Basile et al., 2020; 127 Reuber et al., 2016). This volcanism produced a marine substrate upon which the Late Jurassic-128 Early Cretaceous carbonate section of the Demerara rise developed (Basile et al., 2020; Reuber 129 130 et al., 2016).

During the Early Cretaceous, Southern Africa and South America started to split off 131 because of the counterclockwise rotation of Africa relative to South America (Mann et al., 1995; 132 Pindell, 1991), resulting in two conjugate margins: the Guinea Rise on the African margin and 133 the Demerara Rise on the South American margin. The drift stage of continental separation 134 started to develop in the southernmost part of the South Atlantic Ocean at around 135 Ma (Late 135 Valanginian-Early Hauterivian) (Hall et al., 2018) and then propagated northward toward the 136 Equatorial Atlantic Ocean. This latter propagation of the South Atlantic rifting resulted in the 137 formation of an erosional unconformity affecting the Guyana-Suriname basin and was 138 139 responsible for the sedimentary hiatus between the upper Aptian and the lower Albian (Yang and Escalona, 2011). The pre-Albian section of the Demerara Rise was inverted and cut by a 140 peneplanation surface with a strong angular unconformity prior to the Early Cretaceous opening 141 of the Equatorial Atlantic seaway (Reuber et al., 2016). 142

The Equatorial Atlantic Ocean opened East of the Demerara Plateau during the Late 143 Albian (see Moulin et al., 2010 and references therein). A kinematic change at 105 Ma (Albian) 144 associated to the northward displacement of the triple junction between Africa, North America 145 and South America plates (Müller and Smith, 1993) induced the formation of numerous E-W 146 147 fracture zones North of the Demerara Plateau (Basile et al., 2013). Between the Albian and the Eocene, drifting between Africa and South America induced a general collapse of the unstable 148 shelf edge and is responsible for the main extensional regime in the Guyana-Suriname basin 149 150 (Yang and Escalona, 2011). This predominantly transcurrent setting may have favored the formation of a large pull-apart basin with a gentle thermal subsidence similar to the conjugated 151 152 Guinean continental margin (Benkhelil et al., 1995).

2.2 Stratigraphy

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2.2.1 Stratigraphy of the Guyana-Suriname basin

The sedimentary infilling of the Guyana-Suriname basin may be divided into four stratigraphic 155 156 sequences that overlay the Jurassic basement. These sequences are termed as follows: S1 (Cretaceous), S2 (Paleogene), S3 (Early-Middle Miocene), S4 (Late Miocene to Pliocene) (Yang 157 and Escalona, 2011). Details on the stratigraphy of the S1 sequence and associated references 158 159 can be found in Yang and Escalona (2011). The S1 sequence starts with the Barremian basal 160 clastic sandstones of the Stabroek Formation overlaid by the nonmarine siliciclastic deposits of the Potoco Formation. These formations are separated from the Albian to Turonian shelf marine 161 162 platform deposits belonging to the Canje Formation by a Late Aptian–Early Albian(?) regional 163 unconformity. The Canje Formation is composed of black shales with average total organic 164 contents (TOC) between 4 and 7%, and reaching 29% in the Demerara Rise in the DSDP Site 165 1259 (Meyers et al., 2006). These formations are then overlaid by the New Amsterdam Formation, a siliciclastic interval interpreted as a coastal environment with sand and lagoonal 166 167 deposits (Lawrence and Coster, 1985). This formation corresponds to reservoirs in the Arapaima-1 well in offshore Guyana (Yang and Escalona, 2011). The top of the Mesozoic is 168 marked by an erosional unconformity overlaid by Paleocene to Eocene carbonate shelf deposits 169 170 of the George Town Formation (Yang and Escalona, 2011).

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2.2.2 Stratigraphy of the Arapaima-1 well

The stratigraphy and sedimentology of Cretaceous sequences of the Arapaima-1 well were reinterpreted by the TOTAL department of stratigraphy (Mourlot, 2018) and are presented in Fig. 2. The Early Cretaceous (pre-Aptian) is characterized by medium to coarse grained

sandstones with common shale and limestone intervals deposited in inner to outer shelf 175 paleoenvironments. These clastic deposits are then separated from Late Albian shelf limestones 176 by an erosional unconformity. These Late Albian shelf limestone are then overlaid by the first 177 sequence of green to grey shales slightly enriched in TOC (1% wt) associated with a lowering of 178 the sedimentation rate (Mourlot, 2018). This sequence is gradually overlain by Cenomanian 179 deposits characterized by alternation of shales and sandstones. A two fold increase in 180 sedimentation rate is noted for the Cenomanian clastic deposits when compared with older Early 181 Cretaceous sediments (Mourlot, 2018). This Cenomanian clastic sequence is then followed by 182 183 Cenomanian middle to outer shelf limestone deposits. The uppermost Cenomanian to Coniacian deposits consist in dark-grey to brown shales interbedded with siltstone and very fine sandstone 184 and are characterized by an increase of organic-content (from 2% wt up to 4% wt, Fig. 2). This 185 sequence can be correlated with the Canje Formation which is also composed of organic rich 186 shales with average TOC between 4 and 7%. During the same period, the Demerara plateau also 187 record an enrichment in Total Organic Carbon, with the TOC concentration reaching up to 29% 188 at DSDP Site 1259 (Meyers et al., 2006). The Santonian to Paleocene sediments are 189 characterized by alternation between medium to coarse sandstones and mainly non organic rich 190 191 shales. The sedimentation rate is much higher during the Santonian, respectively six fold and three fold than the Early Cretaceous-Late Albian and Cenomanian-Coniacian intervals 192 (Mourlot, 2018). 193

194 2.3. Sedi

2.3. Sedimentary sources

The potential sources of the studied sediments include the Guiana Shield, the proto Andes, and the Caribbean Large Igneous Province magmatic rocks. Additionally, Aptian-Albian volcanism encountered on the Guinea Plateau (Olyphant et al., 2017) may be a potential source.

198 The Albian volcaniclastic units were dated at *ca* 106 Ma (Olyphant et al., 2017) and related to the silica under-saturated alkaline-nepheline-syenite suite of the Los Archipelago ring structure 199 dated at ca 104.3 ± 1.7 Ma and located off the south-western shore of Conakry, Guinea (Moreau 200 et al., 1996). The Guiana Shield is mainly composed by Paleoproterozoic magmatic and 201 metamorphic rocks accreted to two Archean nuclei during the main or late phase of the Trans-202 203 Amazonian orogeny (i.e., the Imataca block to the northwest and the Amapá block to the southeast; AAIG in Fig.3) (Delor et al. 2003; da Rosa-Costa et al., 2006; Gaudette et al., 1996, 204 1978; McReath and Faraco, 2005; Vanderhaeghe et al., 1998). Based on geochronological and 205 206 structural data, the Guiana shield may be divided into six main provinces decreasing in age southwestward from 2.5 to 1.2 Ga: Amazônia Central, Maroni-Itacaiúnas, Tapajós-Ventuari, Rio 207 Negro-Juruena, Rondônia-San Ignacio, and Sunsás (Santos et al., 2000; Teixeira et al., 1989; 208 Cordani et al. 2000). From a lithological point of view, the northernmost part of the shield is 209 occupied by an extended greenstone belt (GB) amalgamated with Paleoproterozoic granitoids, 210 gneiss, and felsic (meta)volcanic series (PGGF) (Barreto et al., 2013; Gruau et al., 1985), and 211 bounded to the south by a high-grade granulite belt (PHGB) (Klaver et al., 2015; de Roever et al. 212 2003). Overall, the rest of the shield is dominated by younger Paleoproterozoic PGGF marked by 213 214 intrusions of Mesoproterozoic granite plutons (MG) in the Rio Negro province (Tassinari 1981; Sato 1998; Heinonen et al., 2012). Paleoproterozoic sediments related to the erosion of the 215 Trans-Amazonian orogen are also present in the northwest part of the shield and consist in 3000 216 217 m thick alternations of sandstones, conglomerates and pyroclastic rocks (PRS) belonging to the Roraima Formation (Priem et al., 1973). All the units cited above are intruded by sparse 218 Paleoproterozoic mafic intrusions (PMI) (i.e., Avanavero dolerites, (Hebeda et al., 1973; Reis et 219

al., 2013) as well as Jurassic mafic dykes and sills (JMD) related to the Central Atlantic
Magmatic Province (CAMP) activity at 200 Ma (De Min et al. 2003; Deckart et al., 2005).

222 As mentioned above, a rifting affected the Guiana shield from the Jurassic to Cretaceous, resulting in the formation of grabens stretching from SW Guyana to northern Brazil and 223 constituting the Takutu basin. Upstream of the paleo-Berbice delta (Erlich and Keens-Dumas 224 2007), this narrow intracratonic basin is especially important as it could represent the main 225 pathway of the Berbice paleodrainage network during the Cretaceous (Figs. 1 and 3) (Lujan and 226 Armbruster, 2011). The infilling of the basin consists first of the 900-1700 m thick tholeiitic 227 basalts of the poorly dated Apoteri volcanic Formation (Crawford et al., 1985). A Jurassic age is 228 commonly ascribed to these basaltic rocks (Berrangé and Dearnley, 1975; Crawford et al., 1985; 229 230 Yang and Escalona, 2011), in association to the volcanic events of the Central Atlantic Magmatic 231 Province (CAMP) with peak activity at 200 Ma (Marzoli et al., 1999). However, two K/Ar 232 radiometric dating on basaltic lava flows yield Early Cretaceous ages at 114 ± 3 Ma and 116 ± 4 233 Ma (Berrangé and Dearnley, 1975) so that the Apoteri volcanic Formation might be younger than previously thought. Overlying the Apoteri volcanic Formation are the poorly dated shales 234 235 and halites of the Manari and Pirara formations followed by the 1600 cm to 3500 m thick Jurassic to Cretaceous shales, sandstone, siltstones, and micritic limestones of the Takutu 236 Formation (Crawford et al., 1985). This formation developed in fluvial and coastal settings. 237 Pyroclastic flows, up to 87% in SiO₂ content, are recorded in the upper part of the Takutu 238 239 Formation (Crawford et al., 1985).

All the geological units of the Guiana Shield defined above present different ages and lithologies (mafic vs. felsic) making them suitable to distinguish in term of major and trace elemental compositions but also 87 Sr/ 86 Sr and ϵ Nd(0) values. To better characterize the sources

of studied Cretaceous-Paleocene sediments, we compiled 568 published Sr and Nd isotope 243 values measured on magmatic and metamorphic rocks representative of six characteristic end-244 members of the Guiana shield (Table 1 and supporting information Table S1 for details and 245 references). By increasing $\epsilon Nd_{(0)}$ value, these latter include the Archean granitoids (AAIG) with 246 median ɛNd₁₀and ⁸⁷Sr/⁸⁶Sr values at -31.4 (standard deviation (S.D.): 9.3, Table 1) and 0.79509 247 (S.D.: 0.335623, Table 1), the Paleoproterozoic granitoids and felsic series (PGGF) at -23.4 248 (S.D.: 8.0, Table 1) and 0.77395 (S.D.: 0.4638016, Table 1) the Paleoproterozoic high-grade belt 249 (PHGB) at -21.2 (S.D.: 6.2, Table 1) and 0.72025 (S.D.: 0.1571444, Table 1), the 250 Mesoproterozoic granitoids (MG) at -18.5 (S.D.: 2.1, Table 1) and 0.77460 (S.D.: 0.05247612, 251 Table 1), the Paleoproterozoic mafic intrusions and greenstone belt (PMI-GB) at -8.9 (S.D.: 7.1, 252 Table 1) and 0.71051 (S.D.: 0.006878231, Table 1), and the Jurassic mafic dykes (JMD) at -0.2 253 254 (S.D.: 3.1, Table 1) and 0.70647 (S.D.: 0. 0.001692758, Table 1 and Fig. 3). For comparison, this database was completed by 47 87 Sr/ 86 Sr and ε Nd₍₀₎ values of later Cretaceous to modern 255 sediments deposited in the Demerara abyssal plain and Demerara Rise (Martin et al., 2012; 256 Schulte et al., 2009; White et al., 1985), as well as 110 Sr-Nd isotopic compositions of 257 suspended particulate matter and dissolved Sr transported by the main tropical rivers of the 258 northeastern part of South America (i.e., Amazon, lower and upper Orinoco, Rio Negro, Maroni, 259 Trombetas, Caroni) (Allègre et al., 1996; Goldstein et al., 1997; Palmer and Edmond, 1992; 260 Rousseau et al., 2019; Santos et al., 2015). 261

262 **3. Materials and Methods**

263 3.1 Sampling

Nineteen Early Cretaceous to Paleocene fine-grained sedimentary rocks were sampled from the Arapaima-1 exploration well (Fig. 1). These fine-grained sedimentary rocks were deposited in shallow marine mixed clastic and carbonates depositional environments. There is uncertainty regarding the limits between the Late Albian and the Cenomanian and between the Santonian and the Late Campanian in the well due to lack of biostratigraphic data. Otherwise, the time resolution of samples is at the sub-stage level (*i.e.*, ~2 to 3 Ma). These samples provide sufficient time resolution to identify long-term trends in sedimentary provenance.

271

3.2 Bulk organic geochemical analysis

The TOC content of sediments and its stable carbon isotopic compositions ($\delta^{13}C_{TOC}$) 272 273 were analysed at the University of Hanyang (South Korea). Precisely weighed aliquots (0.5-16 g) of crushed and homogenized samples were decalcified by adding about 8 mL of 1M HCl. After 274 shaking overnight (~12 h) to remove carbonates prior to analysis, the TOC content and $\delta^{13}C_{TOC}$ 275 values were determined by EA-IRMS (elemental analyzer-isotope ratio mass spectrometry, 276 EuroEA-Isoprime IRMS, GV instruments, UK). The TOC content and isotope values were 277 calibrated against a standard (IAEA-CH-6, TOC = 42.1 %, δ^{13} C = -10.44 ‰). The analyses were 278 performed at least in duplicate. The TOC data are given in weight percentage (wt%) and the 279 $\delta^{13}C_{TOC}$ values are reported in the standard delta notation relative to the Vienna Pee Dee 280 281 Belemnite (VPDB) standard. The analytical precision (as standard deviation for repeated measurements of the internal standard IAEA-CH-6, n=18) is 1.1 wt% for TOC and 0.2 ‰ for 282 $\delta^{13}C_{TOC}$. 283

3.3 Major and trace elements and Sr-Nd isotopes

285 *3.3.1 Sample preparation*

Each sample was finely crushed in an agate mortar. About 3 g of the crushed sediment 286 were placed into 50 ml centrifuge tubes for sequential leaching to remove the non-terrigenous 287 sedimentary components following the method of Bayon et al. (2002). Three different solutions 288 composed of 5% acetic acid, 15% acetic acid and hydroxylamine hydrochloride (0.05 M) and 289 hydrogen peroxide (H₂O₂, 5%) were then added to remove the carbonates, Fe-Mn oxides and 290 organic carbon, respectively. After the removal of non-terrigenous components, the residual 291 292 fractions were cleaned with ultrapure water (MQ-H₂O, 18 MΩ). Clayey ($<2 \mu$ m) and silty (2-63) µm) fractions were then separated by centrifugation in two steps. First, 25 ml of MQ-H₂O were 293 added to the detrital residues in the tubes, shaken vigorously, and then centrifuged for 2 min at 294 1000 rpm (133 g). The clay-rich supernatants were immediately transferred into new 50 ml 295 centrifuge tubes. Another 25 ml of MQ-H₂O was added to silt-rich detrital residues, mixed 296 thoroughly again, centrifuged for 2.5 min at 800 rpm (85 g), and transferred into corresponding 297 centrifuge tubes. Finally, clay-size fractions were collected after decantation (48 hours) and 298 centrifugation at 3500 rpm (1630 g) (Bayon et al., 2015). These clay-size fractions were then 299 300 analyzed for their major and trace element concentrations and their Sr-Nd isotopic compositions.

301

3.3.2 Major and trace element analyses

The major and trace element concentrations were measured at the Service d'Analyse des Roches et Minéraux (SARM, INSU facility, Vandoeuvre-Les-Nancy, France) by ICP-OES (Na, Mg, K, Ca, Sc, Ti, Mn, Fe, Al, Si and P) and ICP-MS (Rb, Cs, Ba, Sr, Th, U, Y, Zr, Nb, Hf, Cr, V, Co, Cu, Ni, Zn, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu and Ta) after alkali fusion. Analytical details are available on http://helium.crpg.cnrs-nancy.fr/SARM/ and in Carignan et al. (2001). Uncertainty is lower than 5% for elements measured by ICP-OES, and lower than 10% for elements measured by ICP-MS (see supplementary material Table S2).

309

3.3.3 Nd-Sr isotopes compositions

310 The Nd-Sr isotopic compositions were measured at Géosciences Environnement Toulouse 311 (GET) in France. About 100 mg of the leached detrital residue fractions were accurately weighed 312 in cleaned Savillex PFA vials and then digested by several hot acid attacks. The analyzed 313 fractions were firstly digested in hydrogen peroxide for 24 hours at ambient temperature, and then digested in HNO₃ for 24 hours at 80°C followed by HF-HNO₃ for 24 hours at 80°C, and, 314 315 finally HCl+HNO₃ for 24 hours at 115°C. Blank tests were performed to estimate the level of contamination induced by the acid digestion, but it was found to be negligible. Aliquots 316 containing about 1000 ng of Sr and Nd were loaded into the ion exchange columns. Strontium 317 and Nd were separated using the Sr-SPEC, TRU-SPEC and LN-SPEC resins (Eichrom). The 318 isotopic values of both elements were measured using a Finnigan Mat 261 thermal ionization 319 mass spectrometer and a Triton Thermal Ionization Mass Spectrometer in dynamic mode during 320 the same runs as performed for clay-size sediments of the DSDP 25 Site 242 analyzed in 321 (Roddaz et al., 2020). During the Nd runs, a ¹⁴⁶Nd/¹⁴⁴Nd ratio of 0.7219 was used to correct the 322 signal for mass fractionation. For each sample, checks were made for the absence of Samarium 323 324 (Sm). The accuracy of the measurements was estimated on the La Jolla standard. Repeated analyses of the La Jolla standard gave a 143 Nd/ 144 Nd ratio of 0.511867 ± 0.000018 (2 S.D, n = 325 12) in agreement with the recommended value of 0.511858 (Lugmair et al., 1983). During the Sr 326 runs, 86 Sr/ 88 Sr = 0.1194 was used to correct the signal for mass fractionation. The accuracy of the 327 ⁸⁷Sr/⁸⁶Sr measurements was checked against the NBS 987 standard (= 0.710240) Hodell et al., 328

2007). Repeated analyses of the NBS 987 standard gave an 87 Sr/ 86 Sr ratio of 0.710231 ± 0.000032 (2SD, n=15) in agreement with the recommended value of Hodell et al. (2007) therefore no instrumental bias needs to be taken into account. Total blanks (acid digestion plus column chemistry) for Nd and Sr were checked by ICP-MS and found to be negligible (<1 per mil) compared to the Nd and Sr amounts loaded onto the columns.

The measured ¹⁴³Nd/¹⁴⁴Nd_{sample} ratios are expressed in epsilon notation as the fractional deviation in parts per 10^4 (units) from ¹⁴³Nd/¹⁴⁴Nd value of the Chondritic Uniform Reservoir (CHUR). This notation is defined as:

337
$$\varepsilon_{\mathrm{Nd}(t)} = [(^{143}\mathrm{Nd}/^{144}\mathrm{Nd})_{\mathrm{sample}(t)} / (^{143}\mathrm{Nd}/^{144}\mathrm{Nd})_{\mathrm{CHUR}(t)} - 1] \times 10^4$$

where *t* indicates the time at which ε_{Nd} is calculated. Here, no time correction is applied (*t* = 0) and the ¹⁴³Nd/¹⁴⁴Nd_{CHUR(0)} = 0.512638 (Jacobsen and Wasserburg, 1980).

340

341 **4. Results**

342 4.1 Organic geochemistry and $\delta^{13}C_{TOC}$ data

The TOC results for the Arapaima-1 well are variable with values ranging from 0.2 to 4.6 wt% through the studied interval (Table 2). The Cenomanian to Santonian sedimentary rocks (AR1-11B to AR1-17B samples) appear to be more enriched in organic matter (TOC>2%) compared with other sedimentary rocks (Table 2 and Fig. 2). The $\delta^{13}C_{TOC}$ of analyzed samples range between -28.4 (Cenomanian, samples AR1-11E and AR1-11F) and -25.0 (Early Turonian, sample AR1- 12A, Table 2). Compared with compilations of long-term Cretaceous trends in NW Tethyan and Pacific $\delta^{13}C_{bulk}$ data (Bodin et al., 2015; Friedrich et al., 2012), our data show similar sharp positive isotopic excursions in the Cenomanian (between samples AR1-9 and AR1-10, Table 2) and at the Cenomanian-Turonian boundary (samples AR1-11E and AR1-11F, Table 2). However, the younger $\delta^{13}C_{TOC}$ data do not fit the long-term Cretaceous evolution of $\delta^{13}C_{bulk}$ data recorded in the NW Tethyan and Pacific (Friedrich et al. 2012).

4.2 Major elements, Large-Ion Lithophile Elements (LILE), High Field Strength
 Elements (HFSE) and Trace Transition Elements (TTE)

Figure 4 shows the elemental concentrations of major elements, LILE, HFSE and TTE of 356 357 samples normalized to the Post Archean Australian Shales (PAAS) concentration (Pourmand et al., 2012; Taylor and McLennan, 1985). As a whole, the elemental composition of the analyzed 358 sample is rather uniform. They are all depleted in MnO, CaO, K₂O, Rb, Cs, Sr, Th, Y, Zr, Nb, Hf 359 360 et Co against PAAS (Table 3 and Fig. 4). With the exception of two Early Cretaceous samples (AR1-1 and AR1-7) and Santonian- Paleocene samples (AR1-21, AR1-23, AR1-24), they are 361 enriched in Cr and V with respect to PAAS (Table 3 and Fig. 4). However, the analyzed samples 362 are more contrasted regarding their concentrations in Na₂O, P₂O₅, Ba and U against PAAS. 363 Overall, Santonian-Paleocene sediments are less concentrated in major and trace elements when 364 compared with older sediments. But, no apparent trend of major depletion or enrichment in 365 elemental concentration is recorded with the age of sediments. 366

It is possible to assess the degree of source-area weathering and the paleoweathering conditions of ancient sediments using the Chemical Index of Alteration (CIA) (Fedo et al., 1995; Nesbitt and Young, 1982). This index measures the weathering intensity of feldspars relative to unaltered protoliths, and is defined as: $CIA = [Al_2O_3/(Al_2O_3+CaO^*+Na_2O+K_2O)]\times100$ (in molar proportions), where CaO* represents the CaO content in the silicate fraction. CIA values for unaltered plagioclase and K-feldspars typical of unaltered upper crustal rocks are approximately equal to 50, whereas higher CIA values represent higher degrees of weathering. For comparison,
the CIA value of the PAAS standard, which is representative of the upper continental crust
composition, is equal to 67 (Taylor and McLennan, 1985).

Here, the CIA values of analyzed samples range from 61 to 89 and are therefore higher than those of the PAAS, except for one sample (AR1-7, CIA~61). Overall, and with the exception of the Coniacian sample AR1-17A (CIA~74), the Early Cretaceous samples (AR1-1, AR1-6, AR1-7) and Cenomanian samples (AR1-9 to AR1-11F) have lower CIA values (70-78, Table3) than younger Turonian to late Campanian samples (CIA \geq 80, Table 3).

Early Cretaceous to Early Santonian samples have Cr/Th ratios higher than PAAS (37.9-10.2, Table 3) whereas younger samples have Cr/Th ratios lower than PAAS. Analyzed samples have Th/Sc ratios ranging between 0.27 and 1.12 (Table 3). With the exception of two Cenomanian samples (AR1-11E and AR1-11F), the Th/Sc ratios is increasing as the age of the samples is becoming younger (Table 3).

386

3874.3 Rare Earth Elements (REE)

To facilitate comparisons with sediments derived from basic or felsic sources (Cullers, 2000) and the PAAS (Pourmand et al., 2012; Taylor and McLennan, 1985), the Eu anomaly is calculated with respect to chondrites: $Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2}$ where _N refers to the chondrite-normalized concentration value (Condie, 1993). The Cerium anomaly (Ω Ce) normalized to PAAS is calculated following (De Baar et al., 1985) : Ω Ce = 2 × (Ce_{sample}/Ce_{PAAS}) / [(La_{sample}/La_{PAAS}) + (Pr_{sample}/Pr_{PAAS})], where X_{sample} is the concentration of samples and X_{PAAS} refers to the concentrations of the PAAS standard.

When normalized against PAAS, most of the analyzed samples have flat REE patterns 395 and variable Eu anomalies (Fig. 5). The Santonian to Paleocene samples are more enriched in 396 Light REE (LREE) and in Middle REE (MREE) than PAAS (Fig. 5). Three Cenomanian 397 samples (AR1-11B, AR1-11D, AR1-11E) show enrichment in Heavy REE (HREE) against 398 LREE and MREE (Fig. 5 and Table 3). The analyzed sample Ce anomalies display a narrow 399 range of values (0.85-1.01) close to that of the PAAS standard (*i.e.*, $\Omega Ce = 1$). The analyzed 400 values are systematically higher than those measured in authigenic smectites and seawater (ΩCe 401 <0.50, De Baar et al., 1985) and they do not show any variation with stratigraphic position. The 402 Eu/Eu* ratios of the analyzed samples range between 1.26 and 0.67 and are higher compared to 403 the PAAS standard (*i.e.*, $Eu/Eu^* = 0.58$). With the exception of one Cenomanian sample (AR1-404 10), there is an overall increase of the Eu anomaly (i.e decrease of the Eu/Eu* ratio) with the age 405 of the sample (Figs 5 and 6 and Table 3). 406

407 4.4 Sr-Nd isotopes

The ⁸⁷Sr/⁸⁶Sr isotopic composition of the samples range from 0.714598 to 0.773011 while 408 their $\varepsilon_{Nd(0)}$ values range from -0.6 to -21.2 (Table 4). Median values for ${}^{87}Sr/{}^{86}Sr$ isotopic 409 compositions and $\varepsilon_{Nd(0)}$ values are 0.736 and -18.8 respectively. There is no correlation between 410 the Sr and Nd isotopic composition of analyzed samples (see Table S3 in supplementary dataset). 411 Overall, the ⁸⁷Sr/⁸⁶Sr ratios of Arapaima-1 well samples can be split into three groups. Samples 412 AR1-1 (Early Cretaceous) to AR1-9 (Cenomanian) and Coniacian samples (AR1-17A and AR1-413 17B) have quite similar low radiogenic ⁸⁷Sr/⁸⁶Sr ratios ranging between 0.721 and 0.733 (Table 414 4). The other Cenomanian and the Turonian samples show strong variation in their ⁸⁷Sr/⁸⁶Sr 415 ratios (0.715-0.761). And overall, if we except the Early Turonian sample (AR1-12A), the 416 youngest late Santonian to Paleocene analyzed samples have the more radiogenic ⁸⁷Sr/⁸⁶Sr ratios 417

of the analyzed samples (0.749-0.775, Table 4 and Fig. 6). Most of the analyzed samples have low radiogenic Nd isotope composition with $\varepsilon_{Nd(0)}$ values generally between -15.8 and -21.2 (Table 4). Only three samples (Early Cretaceous AR1-1, Cenomanian AR1-11D and Late Campanian AR1-23) have high radiogenic signatures with $\varepsilon_{Nd(0)}$ values between -2.2 and -0.6 (Table 4). Two samples (Cenomanian AR11E and Santonian? AR1-21) have intermediate Nd isotopic composition with $\varepsilon_{Nd(0)}$ values of -12.2 and -13.6 respectively (Table 4).

424 **5. Discussion**

5.1. Influence of weathering, authigenesis and anoxic conditions on provenance proxies 425 The REE content, Eu anomalies, elemental ratios (Cr/Th and Th/Sc) and Sr-Nd isotopic 426 compositions of sedimentary rocks may be used as robust tools for determining the provenance 427 of marine sediments (McLennan et al., 1993), to the condition that they are not affected by 428 weathering and syndepositional clay authigenesis. For instance, high Eu anomalies and Th/Sc 429 and low Cr/Th are characteristic of felsic and more highly differentiated source rocks, whereas 430 high Cr/Th and low Eu anomalies and Th/Sc suggest more mafic and less differentiated source 431 rocks (Cullers, 2000; McLennan et al., 1993b). In general, these ratios as well as the REE 432 content are not significantly affected by diagenesis or weathering (Cullers, 2000; Fedo et al., 433 1995; McLennan, 1989; McLennan et al., 1993a) The absence of any correlation (see Table S3 in 434 supplementary dataset) of CIA and Al/Si with REE content, Eu/Eu*, Cr/Th, Th/Sc, ⁸⁷Sr/⁸⁶Sr 435 isotopic compositions and $\varepsilon_{\rm Nd(0)}$ suggests that chemical weathering has not modified these 436 provenance proxies. Regarding authigenesis effects, any influence from carbonates, Fe-Mn 437 oxides or organic matter formed under marine conditions should have been removed by the 438 sequential carbonate dissolution and leaching steps. In addition, there is no correlation between 439

TOC content and $\delta^{13}C_{TOC}$ and provenance proxies (Eu/Eu*, Cr/Th, Th/Sc, ${}^{87}Sr/{}^{86}Sr$ isotopic 440 compositions and $\varepsilon_{\rm Nd(0)}$ suggesting that enrichment and preservation of organic matter are not 441 affecting these proxies. Moreover, the REE patterns of authigenic smectites formed in deep-442 water environments are characterized by LREE depletion, HREE enrichment and strong negative 443 Ce anomalies (Ω Ce < 0.50, De Baar et al., 1985) typical of seawater (Piper, 1974). Here, most of 444 the analyzed samples have Ce anomalies higher than or equal to 0.85, with flat REE patterns 445 against PAAS corresponding to the "shales" field in Fig. 7. Only three Cenomanian samples 446 (AR1-11B, AR1-11D, AR1-11E) show enrichment in HREE over LREE and plot between the 447 "shales" and "seawater" fields in Fig. 7. However, as they have low Ce anomalies against PAAS 448 449 (Ω Ce between 0.88 and 0.92), this suggests that, even if authigenic smectites might have been incorporated during early diagenetic processes, its proportion was much less than the detrital 450 fraction. Finally, it is worth noting that diagenetic remobilization of Eu under reducing 451 conditions might lead to the development of high positive Eu anomalies (MacRae et al., 1992). 452 As anoxic to suboxic conditions are suggested from geochemical and sedimentological proxies 453 onto the Demerara Rise (i.e., Ocean Drilling Program Leg 207 Site) in the Cenomanian-Turonian 454 interval (Berrocoso et al., 2008; Hetzel et al., 2009), this process could explain the high Eu/Eu* 455 value (1.26) of the Cenomanian sample AR1-10 (Table 3 and Fig.6). However, anoxic conditions 456 are also known to favor organic matter preservation and can be recorded by high TOC content 457 (Algeo and Liu, 2020; Schlanger and Jenkyns, 1976), which is not the case for the AR1-10 458 sample that has one of the lowest TOC content among the analyzed samples (i.e. 0.5%, Table 2). 459 Rather, the samples with the highest TOC content (TOC>2%, Table 1) have lowest Eu/Eu* ratios 460 (0.72-0.88, Table 3) suggesting that diagenetic remobilization of Eu under reducing condition 461 was negligible in our samples. 462

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5.2. Provenance interpretation

Most of the $\delta^{13}C_{TOC}$ values of analyzed samples have organic matter mainly of terrestrial origin because they have $\delta^{13}C_{TOC}$ values within the range of C3 plant-derivated organic matter (-29.3 to -25.5‰, mean~27‰ (e.g. (Tyson, 1995) and depart from typical marine $\delta^{13}C_{TOC}$ values (-18 to-22‰ (e.g.(Meyers, 1997)). Hence, this suggests that the analyzed sedimentary rocks were mainly fed by continental detritus.

The potential sources of the studied sediments may include the rocks of the Guiana Shield, 470 471 Northwestern Africa, the proto Andes and the Caribbean Large Igneous Province magmatic rocks. Here, we argue that a northwestern African provenance is unlikely because existence of 472 bottom currents parallel to the northwestern African coast during the Cretaceous (Mourlot et al., 473 2018b) may have prevented the arrival of northwestern African detritus to the deep distal part of 474 the Central Atlantic Ocean (Mourlot et al., 2018b). In addition, aerial transport from northwest 475 Africa is also unlikely because $\varepsilon_{Nd(0)}$ values of our analyzed sedimentary rocks and those of the 476 Demerara rise (Martin et al., 2010) show significant differences with those of contemporaneous 477 sedimentary rocks deposited in the deep offshore Central Atlantic Ocean and northwestern 478 African margin during the Albian-Maastrichtian times (Mourlot et al., 2018a). A proto-Andean 479 provenance is not supported by the provenance of Cretaceous sedimentary rocks deposited in the 480 Amazonian retroarc foreland basin because these sedimentary rocks are cratonic in provenance 481 and were deposited by cratonic paleodrainage (Erlich et al., 2018; George et al., 2019; Hurtado et 482 al., 2018; Louterbach et al., 2018) with very negative $\varepsilon_{\rm Nd(0)}$ values (Hurtado et al., 2018; 483 Louterbach et al., 2018) originated from the Central part of the Amazon Shield (Hurtado et al., 484 2018). 485

When plotted on a ⁸⁷Sr/⁸⁶Sr vs. $\varepsilon_{Nd(0)}$ diagram (Fig. 8) and compared with relevant end-members, including the Precambrian units of the Guiana Shield, the Jurassic mafic dykes related to the CAMP, marine sediments of the Demerara rise and abyssal plain and Suspended Particulate Matters (SPM) transported by modern rivers draining the northern part of South America and the Andes, three groups can be distinguished based on their differences in $\varepsilon_{Nd(0)}$ values.

The first group includes the samples AR1-1 (Early Cretaceous), AR1-9 (Cenomanian) 491 and AR1-23 (Late Campanian) and is characterized by $\varepsilon_{Nd(0)}$ values >-2.2 close to the mean $\varepsilon_{Nd(0)}$ 492 value of the Jurassic mafic dykes (JMD) (i.e. mean= 0.4 Standard Deviation= 3.1, Table S1 493 (Deckart et al., 2005; Min et al., 2013) and close to Precambrian Avanavero Mafic Intrusions 494 (PMI) (Hebeda et al., 1973; Leal et al., 2012), and some tholeiitic and komatiitic rocks of the 495 Paleoproterozic greenstone belts (GB) (Gruau et al., 1985; Kaminsky et al., 2004; McReath and 496 Faraco, 2005; Voicu et al., 2000) (Figs. 3 and 9). Hence, these high $\varepsilon_{Nd(0)}$ values could indicate 497 provenance from Precambrian or Jurassic mafic source outcropping in the northernmost parts of 498 the Guiana Shield. Alternatively, these high $\varepsilon_{\rm Nd(0)}$ values could also attest to the presence of 499 active basic volcanism during the deposition of analyzed samples. 500

The two oldest Early Cretaceous (AR1-1) and Cenomanian (AR1-9) samples have trace 501 elements ratios typical of "basic" sediments (i.e., sediments derived from a basic source sensu 502 Cullers (2000)), with Eu/Eu* >0.70, Th/Sc<0.4 and Cr/Th > 22.2 (Table 3 and Fig. 6). From a 503 geochemical point of view, it is not possible to distinguish between the Precambrian or Jurassic 504 505 mafic rocks outcropping in the Guiana Shield and basic volcanism originated from the CLIP, Bahamas hot spot or Apoteri volcanism from the Takutu rift. The AR1-9 sample is marked by a 506 positive $\varepsilon_{Nd(0)}$ excursions of +17.8 to 18.2 units when compared with overlying AR1-8 and 507 underlying AR1-10 samples respectively (Fig. 6 and Table 4). We believe that these positive 508

 $\varepsilon_{\rm Nd(0)}$ excursions sample cannot be explained by a contribution of Precambrian or Jurassic mafic 509 510 rocks of the Guiana Shield and may favor a basic volcanic source for AR1-9 sample. In effect, Precambrian and Jurassic mafic rocks of the Guiana Shield always outcrop as patches in more 511 512 differentiated magmatic rocks (Fig. 3) and it is likely that admixture of sediments derived from mafic and more differentiated sources would have resulted in a more negative $\varepsilon_{\rm Nd(0)}$ value for 513 AR1-9 sample. In addition, if mafic rocks outcropping in the Guiana Shield would have been 514 515 eroded alone, the process leading to the erosion of these rocks would have to be progressive and 516 this would be reflected by a progressive shift toward the highest $\varepsilon Nd(0)$ values and not a positive $\varepsilon_{\rm Nd(0)}$ excursions of +17.8 to 18.2 units. Hence, we favor a Cenomanian basaltic volcanic source 517 for sample AR1-9. Potential candidates may include the basic volcanism of the Caribbean Large 518 Igneous Province (CLIP) active between 94 and 63 Ma (Loewen et al., 2013), or the basic 519 520 volcanism of the Apoteri volcanic Formation outcropping in the Takutu rift (Crawford et al., 1985). Regarding the latter potential source, Proterozoic or Jurassic ages are generally ascribed 521 to the basaltic flows (Crawford et al., 1985). However, two K/Ar dating measured on basaltic 522 523 lava flows yield Early Cretaceous ages at 114 ± 3 Ma and 116 ± 4 Ma (Berrangé and Dearnley, 1975,) so that a Cretaceous age for the Apoteri basaltic flow might be possible. However, a 524 volcanic basaltic source might not be true for the oldest Early Cretaceous sample AR1-1. This 525 sample is the only sample analyzed below the Late Albian unconformity (Figs 2 and 6) and 526 527 because of this low sampling resolution, it is not possible to discard a progressive shift of $\varepsilon Nd(0)$ values. Hence, both contemporaneous basic volcanic rocks and mafic rocks of the Guiana Shield 528 are possible sources for this sample. Potential volcanic source for this sample include the basic 529 530 volcanism from the CLIP or the Apoteri Formation as well as Aptian-Albian volcanism found on the Guinea Plateau (Olyphant et al., 2017). 531

The Late Campanian sample (AR1-23) has trace element ratios characteristics of felsic 532 sediments sensu (Cullers, 2000) with Eu/Eu* <0.70, Th/Sc>0.64 and Cr/Th < 14.9 (Table 3 and 533 Fig. 6). These exclude mafic sources or basic volcanism as potential sources for this sample and 534 suggest an origin linked to a more acid volcanic activity. The only regional source for this acid 535 volcanism would be the pyroclastic flow recorded in the upper part of the Takutu Formation of 536 the Takutu basin (Crawford et al., 1985). These pyroclastic flows have up to 87% of SiO₂ 537 content but have never been dated. In fact, there is no radiometric nor biostratigraphic dating for 538 the Takutu Formation and for the underlying and overlying formations. Even the positions of the 539 Jurassic/Cretaceous and Cretaceous/Cenozoic boundaries are not constrained. Hence, a Late 540 Cretaceous age for the Takutu Formation cannot be excluded and, in the absence of other option, 541 the pyroclastic flows mentioned at the top of the formation could be the source of the Late 542 Campanian sample. 543

The second group consists of AR1-11E (Cenomanian) and AR1-21 (Santonian-544 Campanian) samples and is characterized by intermediate $\varepsilon_{Nd(0)}$ values (-12.2 and - 13.6 545 respectively, Figs. 6 and 8 and Table 4). In the case of Amazonian sediments, this $\varepsilon_{Nd(0)}$ value is 546 generally used as a cutoff value for identifying more radiogenic Andean-derived sediments 547 (Figueiredo et al., 2009; Horbe et al., 2014; Hurtado et al., 2018; Roddaz et al., 2005). The Sr-Nd 548 549 isotopic composition of Amazonian Andean sediments is commonly interpreted to reflect mixing between two end-members: the juvenile primitive Andean Arc and the older upper continental 550 crust of the Brazilian shield (Basu et al., 1990; Roddaz et al., 2005). Applying this finding to our 551 results suggests that the two analyzed samples with intermediate $\varepsilon_{Nd(0)}$ values might result from 552 mixing between the northern mafic sources listed above (i.e., JMD, PMI and GB) or basic 553 volcanic sources (CLIP, Bahamas hot spot or Apoteri volcanism) and the differentiated 554

Precambrian granitoids and metamorphic rocks (e.g., PGGF, PHGB) (Fig. 8). However, more
 samples are needed to test a mixing hyperbole between these two end-members.

The third group of samples encompasses all other analyzed and is characterized by $\varepsilon_{\rm Nd(0)}$ 557 values inferior to -15.8 with a median $\varepsilon_{Nd(0)}$ value at -19.2 (Table 4, Fig. 8). Such low $\varepsilon_{Nd(0)}$ 558 values are typical of Amazonian cratonic-derived sedimentary rocks and present-day cratonic-559 derived river sediments (e.g., Maroni, upper Orinoco, Trombetas rivers) dominated by supplies 560 561 from Paleoproterozoic and Mesoproterozoic granites, gneiss and metamorphic series (PFFG) covering the more inner parts of the craton (Figs. 3 and 8) (Allègre et al., 1996; Goldstein et al. 562 1997; Bayon et al., 2015; Figueiredo et al., 2009; Roddaz et al., 2005). For instance, Suspended 563 564 Particulate Matter (SPM) transported by the Maroni River draining the Paleoproterozoic terranes to the east of the Guiana shield have $\varepsilon_{Nd(0)}$ values between -21 and -25.2 (Rousseau et al., 2019) 565 (Fig. 3 and 8). Compared with the modern Maroni SPM, the Cretaceous sediments from the 566 Guyana-Suriname basin have a slightly more juvenile Nd isotopic ratios but they have similar 567 $\varepsilon_{\rm Nd(0)}$ values as the Upper Orinoco river fine-grained sediments analyzed in Goldstein et al. 1997 568 (-19.6 and -20.5 respectively, Table S1) suggesting a similar origin (Fig. 3). 569

Based on their $\varepsilon_{\rm Nd(0)}$ values, most of the analyzed samples derive from the Guiana Shield 570 with variable contributions from two main sources: 1) Paleoproterozoic granitic and 571 metamorphic units covering the innermost parts of the shield and 2) Jurassic or Precambrian 572 mafic dykes and greenstone belts. More insight can be gained with trace elements ratios and 573 ⁸⁷Sr/⁸⁶Sr isotopic ratios. For instance, the four Late Santonian to Paleocene samples have more 574 radiogenic ⁸⁷Sr/⁸⁶Sr ratios than the oldest samples. These younger samples have Eu/Eu*, Th/Sc 575 and Cr/Th ratios close to that of the PAAS whereas oldest samples have Eu/Eu*, Th/Sc and 576 Cr/Th ratios close to those defining "basic sediments" sensu Cullers (2000) (Fig. 6). As there is 577

578 no coeval change in $\varepsilon_{Nd(0)}$ values, these temporal changes in trace elements ratios and ⁸⁷Sr/⁸⁶Sr 579 isotopic ratios could result from increasing contributions of more felsic sources rather than 580 contribution from older sources. Potential candidates for these felsic sources could be 581 Paleoproterozoic granitoids, gneiss and felsic volcanic units, Mezoproterozoic granites, or high-582 grade granulite belts (PGGF, MG, PHGB in Fig. 3 respectively). Both units outcrop in the inner 583 part of the Guiana Shield and thus imply an extension of the drainage area toward the South 584 between the Coniacian and the Santonian (Fig. 9).

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5.3 Implications for the Late Cretaceous evolution of Atlantic equatorial margins

Our geochemical data support that Early Cretaceous-Paleocene sediments of the Guyana-586 587 Suriname basin have a Guiana cratonic provenance. Analysis of 2D seismic lines suggest that the deep offshore part of the Guyana-Suriname basin was supplied by these cratonic sediments via a 588 large canyon system fed by the proto-Berbice river (Figs.1 and 9) with a paleo-delta probably 589 590 located between the present-day Berbice and Corantijn rivers (Erlich and Keens-Dumas 2007; Mourlot, 2018; Yang and Escalona, 2011). Our provenance data also suggests an extension of 591 the proto-Berbice drainage area toward the inner parts of the Guiana craton after the Coniacian 592 (~86 Ma, Fig. 9), likely through the Takutu rift. This provenance change was associated with a 593 decrease in TOC content and a six fold increase in sedimentation rate in the Arapaima-1 well 594 (Mourlot, 2018), suggesting increasing erosion of inner parts of the Guiana Shield. 595 Geomorphological and thermochronological data also attest to uplift and erosion of the inner 596 domains of the Guiana Shield during the Late Cretaceous (Sapin et al., 2016). In the easternmost 597 598 part of Suriname and in Brazil (close to the Brazilian-Suriname frontier) (see Fig. 7 in Sapin et al., 2016), a Late Cretaceous erosive surface ("peneplanation surface") termed S0 exposed at 599 elevation $> \sim 900$ m above sea level (asl) has been identified and mapped (Sapin et al., 2016). 600

This surface was dated by Ar/Ar on lateritic material at ~71 Ma and Apatite Fission Track 601 Analysis at 77 ± 3 Ma (Sapin et al., 2016 and references therein) suggesting that a significant 602 erosion affected this part of the craton during the Campanian. This S0 surface outcrops 603 immediately above the PGGF basement and hence has recorded the uplift and erosion of this 604 terrane, which would be consistent with the increasing contribution of a felsic source to the Late 605 Santonian-Paleocene sediments. This post Coniacian uplift of the Guiana Shield post-dated by 606 >20 Ma the post-rift Albian tectonic inversion ascribed to a plate kinematic change that affected 607 the Demerara Plateau (offshore French Guiana) at ca 105 Ma (Basile et al., 2013). In 608 northeastern Brazil, various studies have presented evidences for a Late Cretaceous uplift 609 (Harman et al., 1998; Morais Neto et al., 2009; Peulvast and Bétard, 2015; Potter, 1997). 610 Increase of denudation at ~80-60 Ma of the eastern Central Brazilian shield is documented by 611 thermal histories derived from fission track data (Harman et al., 1998). This uplift and erosion of 612 613 the Central Brazilian shield is also attested by the development of the high elevated (~720 m asl) 614 Brazilian Carajas surface in northeastern Amazonia which started its weathering history at ca. 80 Ma (Monteiro et al., 2018). Late Cretaceous fission track cooling ages (Morais Neto et al., 2009) 615 616 as well as presence of Cenomanian marine deposits associated to development of Paleogene 617 laterites on the top of the Borborema Plateau (Peulvast and Bétard, 2015) indicate that the 618 Borborema plateau (Northeastern Brazil) underwent uplift and denudation between the 619 Cenomanian and Early Cenozoic. In the conjugate margin of NW Africa, the Campanian–Maastrichtian is characterized by increasing input of Precambrian detritus along the 620 621 Northwest African Margin and a progression of drainage areas toward the east and the inner units of the West African Craton (Mourlot et al., 2018a). 622

Based on this literature review, both the northeastern South American and West African 623 cratons experienced coeval post Coniacian uplift of their inner parts suggesting that a similar 624 post-rift geodynamic mechanism occurred during the opening of the Atlantic Equatorial Ocean. 625 This coeval uplift of the Atlantic margins could result from significant intracontinental 626 deformation caused by major change in the relative plate motions between Africa, Antarctica, 627 and South America that began between anomaly C34 (83 Ma) and C31 (67 Ma) (see Harman et 628 al., 1998 and references therein) as well as mantle upwelling under the margin (Matton and 629 Jébrak, 2009) or a combination of these. Furthermore, given the presence of a volcanic eruption 630 631 in the Late Campanian as recorded by the sample AR1-23, the presence of an active plume mantle upwelling cannot be discarded. 632

633 **6. Conclusions**

Based on the TOC content, $\delta^{13}C_{TOC}$ data, major and trace element concentrations and Sr– Nd isotopic compositions of Cretaceous to Paleocene sedimentary rocks sampled in the Arapaima-1 well (Guyana-Suriname basin) compared with an extensive Sr-Nd isotopic composition of possible sources, this provenance study yields the following conclusions:

Three samples AR1-1 ((Early Cretaceous, AR1-9 (Cenomanian,) and AR1-23 (Late 638 • Campanian in age) have $\varepsilon_{Nd(0)}$ values >-2.2 that might have recorded volcanic activity 639 during their deposition. However, this interpretation might not be true for the oldest 640 sample AR1-1 that could also be sourced by mafic rocks from the Guiana Shield. The 641 Cenomanian sample may have recorded basic volcanic activity of the Caribbean Large 642 Igneous Province or the Takutu rift, whereas the Late Campanian sample would record 643 acid volcanic eruption associated with the pyroclastic flow having affected the Takutu rift 644 in the Cretaceous; 645

All other analyzed samples have an overall Guiana cratonic provenance with variable 646 contributions from Paleoproterozoic and Mesoproterozoic and greenstone belt, Jurassic 647 and Precambrian mafic dykes. Two Cenomanian and Santonian-Campanian samples 648 (AR1-11F and AR1-21) have intermediate $\varepsilon_{Nd(0)}$ values of ~ -13 suggesting more 649 important contribution of mafic rocks from the Guiana Shield or basic volcanic rocks 650 from the CLIP, Bahamas hot spot or Apoteri volcanism. The other samples have $\varepsilon_{_{Nd/0}}$ <-651 15.8 and suggest a dominant provenance from the inner Paleoproterozoic granitic units of 652 the Guiana Shield; 653

• Late Santonian to Paleocene samples have more radiogenic ⁸⁷Sr/⁸⁶Sr isotopic ratios and trace element ratios characteristic of more differentiated sources when compared with oldest samples. This suggests an increasing contribution from the Paleoproterozoic granitoids, gneiss and felsic volcanism units or the Mesoproterozoic granites through time. This also implies an evolution of the drainage area toward the inner parts of the Guiana Shield between the Coniacian and the Santonian.

660 This post-Coniacian erosion of the inner parts of the Guiana Shield is recorded in various 661 other parts of the northeastern South American and West African cratons. This suggests that 662 they experienced a similar uplift mechanism occurring during the opening of the Atlantic 663 Equatorial Ocean. Probable causes could be a major change in the relative plate motions 664 between Africa and South America as well as post rift mantle upwelling under the margin. 665 The latter hypothesis could be favored by the supposed presence of active volcanism during 666 the deposition of the analyzed samples.

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677 **Data availability**

All geochemical data including total organic carbon (TOC) content, $\delta^{13}C_{TOC}$, major and trace 678 element concentrations, and Sr-Nd isotopic data (Table 2 to 4) as well as compiled database, 679 major and trace element uncertainties and correlation coefficients (Table S1 to S3) are uploaded 680 into an online supplementary dataset associated with this paper (Roddaz, Martin; Dera, 681 Guillaume; Mourlot, Yannick; Calves, Gerome; Kim, Jung-Hyun; Chaboureau, Anne-Claire; 682 Mounic, Stephanie; Raisson, Francois (2020), "Geochemistry of Arapaima well sediments 683 Roddaz et al. in Revision to Marine Geology", Mendeley Data, V2, doi: 10.17632/jnwgsn98tt.2, 684 685 http://dx.doi.org/10.17632/jnwgsn98tt.2)).

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1225 Figure captions

Figure 1. A) Location of study area in its Cretaceous paleogeographical context (Torsvik et al.,
2012). The red rectangle indicates position of area shown on Figure 1B. WAC: West African
Craton. B) Map of the Precambrian Guiana Shield, Mesozoic Takutu basin (TkB), and location
of studied Arapaima-1 well (AR-1, red circle)

Figure 2. Stratigraphy and sedimentary log section of Arapaima-1 well including $\delta^{13}C_{TOC}$ data, 1230 1231 TOC contents, sedimentation rate and main geodynamic events at the scale of Guyana-Suriname basin. The stratigraphic position of analyzed samples is indicated by orange triangle. AR-1: 1232 1233 Arapaima-1 well. M: mudstone. W: wackestone. P: packstone. G: grainstone. C: clay. S: silt. FS: 1234 fine sandstone. MS: medium sandstone. CS: coarse sandstone. Details on the calculation of 1235 sedimentation rates can be found in (Mourlot, 2018). References for timing and duration of main 1236 geodynamic events are as follow : South Atlantic Ocean opening (Hall et al., 2018), Equatorial Atlantic Ocean opening (Moulin et al., 2010; Sibuet and Mascle, 1978), Albian-Coniacian 1237 1238 cooling (Benkhelil et al., 1995; Yang and Escalona, 2011), Central Atlantic spreading rate acceleration (Larson, 1991), Caribbean Large Igneous Province (Loewen et al., 2013), Andean 1239 deformation (Hurtado et al., 2018; Louterbach et al., 2018). 1240

Figure 3. Map showing the main geological units of the Guiana Shield (simplified from (Kroonenberg et al., 2016)) and the localization of the main northern South American rivers. Mean values for $\varepsilon_{Nd(0)}$ and ${}^{87}Sr/{}^{86}Sr$ isotopic composition of the main geological unit of the Guiana Shield are given (See Table 1 and text for details and references).

Figure 4. Major and selected trace element contents of analyzed samples normalized to PAAS
(Pourmand et al., 2012; Taylor and McLennan, 1985). From left to right on the diagram, these

include the major elements (SiO₂, TiO₂, Al₂O₃, Fe₂O₃, MnO, MgO, CaO, Na₂O, K₂O and P₂O₅),

1248 LILE (Rb, Cs, Ba, Sr, Th and U), HFSE (Y, Zr, Nb, Hf), and TTE (Sc, Cr, Co, V and Ni).

Figure 5. Rare earth element (REE) contents of studied samples normalized to PAAS (Pourmand
et al., 2012; Taylor and McLennan, 1985).

Figure 6. Stratigraphic variation of Nd and Sr isotope compositions (expressed as $\mathcal{E}_{Nd(0)}$ and 1251 ⁸⁷Sr/⁸⁶Sr) and selected trace element ratios (Cr/Th, Th/Sc and Eu/Eu*, see text for details) of 1252 1253 analyzed samples. The lengths of the horizontal bar for the Sr-Nd isotopic compositions 1254 correspond to 1_o analytical errors (see Table 4). The horizontal bars for the Cr/Th, Th/Sc, and 1255 Eu/Eu* ratios correspond to 15% (i.e., the sum of the Cr and Th concentration analytical errors), 25% (i.e., the sum of the Th and Sc concentration analytical errors) and 5% (i.e., the Eu 1256 1257 concentration analytical error, see supplementary material Table S2). Dashed lines in Eu/Eu*, Th/Sc and Cr/Th show the PAAS ratio values (Taylor and McLennan, 1985; Pourmand et al., 1258 1259 2012). Red and blue rectangles indicate the values range for felsic and basic sediments, respectively (Cullers, 2000). 1260

Figure 7. HREE/LREE vs. MREE/MREE* diagram. HREE/LREE and MREE/MREE* values are calculated following (Martin et al., 2010) where HREE=Tm+Yb+Lu, LREE=La+Pr+Nd, MREE=Gd+Tb+Dy (all PAAS normalized), and MREE*=the average of HREE+LREE. The MREE bulge and "bell-shaped" REE profiles correspond to REE patterns observed in fish teeth, Fe-Mn oxides, organic matter and pore waters, while "HREE-enriched" profiles correspond to modern sea water (Huck et al., 2016; Moiroud et al., 2016). These end-members reflect the REE contents of marine sediments influenced by seawater or authigenic phases, while "flat" REE

patterns are characteristic of continental clays (see Huck et al., 2016; Moiroud et al., 2016 for a
review). Analyzed samples are represented by specific symbols used in Figs 4 and 5.

Figure 8. Cross-plots of $\varepsilon_{Nd(0)}$ and ${}^{87}Sr/{}^{86}Sr$ values for the analyzed samples. The isotopic fields 1270 of potential end-members of the Guiana Shield are based on a compilation of published data (see 1271 Table 1 and supporting Table S1). AAIG: Archean granitoids (AAIG) (Barreto et al., 2013; da 1272 Rosa-Costa et al., 2006; Hurley et al., 1968; Montgomery and Hurley, 1978; Oliveira et al., 1273 1274 2008; Olszewski et al., 1989; Othman et al., 1984; Posadas and Kalliokoski, 1967; Tassinari et 1275 al., 2004) (Avelar et al. (2003); PGGF: Paleoproterozoic granitoids and felsic series (Barreto et al., 2013; Boher et al., 1992; da Rosa-Costa et al., 2006; Gaudette et al., 1978; Gruau et al., 1985; 1276 1277 Hurley et al., 1968; Lenharo et al., 2002; Marques et al., 2014; McReath and Faraco, 2005; Nogueira et al., 2000; Priem et al., 1973, 1971; Sato, 1998; Tassinari, 1981; Voicu et al., 2000), 1278 Macambira et al. (1987) Jorge Joao et al. (1985), Gibbs and Barron (1993 Avelar et al. (2003); 1279 1280 PHGB: Paleoproterozoic high-grade belt (Gaudette et al., 1996; Heinonen et al., 2012; Klaver et al., 2015; Othman et al., 1984; Sato, 1998; Spooner et al., 1971; Tassinari, 1981) de Roever et al. 1281 (2003); MG: Mesoproterozoic granitoids (Heinonen et al., 2012; Souza et al., 2015; Tassinari, 1282 1981); PMI-GB: Paleoproterozoic mafic intrusions and greenstone belt (Gibbs et al., 1986; 1283 Gruau et al., 1985; Hebeda et al., 1973; Kaminsky et al., 2004; Leal et al., 2006; McReath and 1284 1285 Faraco, 2005; Nixon et al., 1992; Santos et al., 2000; Sato, 1998; Voicu et al., 2000); JMD: Jurassic mafic dykes (Deckart et al., 2005; Min et al., 2013). To avoid outlier effects and 1286 important overlapping, the limits of each rectangular field are defined as the first quartile and the 1287 1288 third quartile for each subset. Comparative clay data from the Demerara Rise and Demerara abyssal plain are added (Martin et al., 2012; Schulte et al., 2009; White et al., 1985). Sr-Nd 1289 isotopic compositions of suspended particulate matter and dissolved Sr transported by the main 1290

tropical rivers of the northeastern part of South America (i.e., Amazon, lower and upper Orinoco,
Rio Negro, Maroni, Trombetas, Caroni) (Allègre et al., 1996; Goldstein et al., 1997; Palmer and
Edmond, 1992; Rousseau et al., 2019; Santos et al., 2015)are added. Note that, when unavailable,
sediment ⁸⁷Sr/⁸⁶Sr values of rivers were replaced by water ⁸⁷Sr/⁸⁶Sr values. **Figure 9**. Paleogeographic map at 90 Ma (modified from Scotese and Golonka (1997) showing

1297 curve) and post-Coniacian (blue dashed curve) times. TkB: Takutu basin. AR-1: Arapaima-1

potential provenance areas for the Guyana-Suriname basin for pre-Coniacian (in red dashed

1298 well.





Precambrian Guiana Shield Mesozoic sedimentary basin

Continent

Shallow marine

Deep marine













AR1 well







 $0.700 \quad 0.710 \quad 0.720 \quad 0.730 \quad 0.740 \quad 0.750 \quad 0.760 \quad 0.770 \quad 0.780 \quad 0.790 \quad 0.800$



٤ _{Nd(0)}	AAIG	PGGF	PMI+GB	MG	PHGB	JMD
Ν	59	93	32	8	31	43
Min	-42.8	-42.8	-19.3	-21.2	-36.4	-4.6
Max	-1.9	-11.4	4.1	-15.3	-2.1	5.7
Mean	-30.2	-23.9	-7.8	-18.3	-20.4	0.4
Std. error	1.2	0.6	1.2	0.8	1.4	0.5
Variance	86.5	38.2	49.9	4.5	64.5	9.4
Stand. dev	9.3	6.2	7.1	2.1	8.0	3.1
Median	-31.4	-23.4	-8.9	-18.5	-21.2	-0.2
25 prcntil	-37.1	-26.6	-14.2	-20.5	-24.1	-2.1
75 prcntil	-23.9	-19.5	-1.9	-16.5	-17.1	3.6

⁸⁷ Sr/ ⁸⁶ Sr	AAIG	PGGF	PMI+GB	MG	PHGB	JMD
Ν	52	214	26	5	40	51
Min	0.70857	0.7015	0.704448	0.7645	0.7012	0.70292
Max	2.7365	4.4008	0.72945	0.8804	1.58175	0.711148
Mean	0.8880397	0.9097801	0.7111302	0.80524	0.7598767	0.706221
Std. error	0.0465425	0.0317048	0.0013489	0.023468	0.0248467	0.000237
Variance	0.1126428	0.2151119	4.73E-05	0.0027537	0.0246944	2.87E-06
Stand. dev	0.335623	0.4638016	0.0068782	0.0524761	0.1571444	0.0016928
Median	0.795095	0.77395	0.71051	0.7746	0.72025	0.70647
25 prcntil	0.7489525	0.738245	0.7048952	0.76535	0.7065225	0.70462
75 prcntil	0.838575	0.875025	0.7157525	0.86045	0.7402675	0.70746

Table 1: Mean ɛNd(0) values and 87Sr/86Sr isotopic composition and statistical parameters for potential end members; N: number of samples analyzed

Std error: Standard error; Stand. Dev: Standard deviation; 25 prcntil: first quartile; 75 prcntil: third quartile;

AAIG: Archean granitoid ; PGGF: Paleoproterozoic granitoids and felsic series; PMI+GB: Paleoproterozoic mafic intrusions and greenstone belt;

PHGB: Paleoproterozoic high-grade belt; MG: Mesoproterozoic granitoids; JMD: Jurassic Mafic Dykes

Data references:

AAIG: Barreto et al., 2013; da Rosa-Costa et al., 2006; Hurley et al., 1968; Montgomery and Hurley, 1978; Oliveira et al., 2008; Olszewski et al., 1989; Othman et al., 1984;

Posadas and Kalliokoski, 1967; Tassinari et al., 2004) (Avelar et al. (2003); PGGF: Paleoproterozoic granitoids and felsic series (Barreto et al., 2013; Boher et al., 1992; da Rosa-Costa et al., 2006; Gaudette et al., 1978; Gruau et al., 1985; Hurley et al., 1968; Lenharo et al., 2002; Marques et al., 2014; McReath and Faraco, 2005; Nogueira et al., 2000; Priem et al., 1973, 1971; Sato, 1998; Tassinari, 1981; Voicu et al., 2000), Macambira et al. (1987) Jorge Joao et al. (1985), Gibbs and Barron (1993 Avelar et al. (2003); PHGB: Paleoproterozoic high-grade belt (Gaudette et al., 1996; Heinonen et al., 2012; Klaver et al., 2015; Othman et al., 1984; Sato, 1998; Spooner et al., 1971; Tassinari, 1981) de Roever et al. (2003) ; MG : Mesoproterozoic granitoids (Heinonen et al., 2012; Souza et al., 2015; Tassinari, 1981); PMI-GB: Paleoproterozoic mafic intrusions and greenstone belt (Gibbs et al., 1986; Gruau et al., 1973; Kaminsky et al., 2004; Leal et al., 2006; McReath and Faraco, 2005; Nixon et al., 1992; Santos et al., 2000; Sato, 1998; Voicu et al., 2000); JMD: Jurassic mafic dykes (Deckart et al., 2005; De Min et al., 2003)

Sample	Age	δ ¹³ C _{TOC} (‰ VPDB)	TOC (wt%)
AR1-1	Early Cretaceous	-26.7	0.2
AR1-6	Aptian	-28.0	1.1
AR1-7	Late Albian	-27.0	0.3
AR1-8	Late Albian	-26.1	0.5
AR1-9	Cenomanian	-28.0	1.0
AR1-10	Cenomanian	-25.8	0.5
AR1-11B	Cenomanian	-27.9	2.0
AR1-11C	Cenomanian	-28.3	1.3
AR1-11D	Cenomanian	-27.7	2.5
AR1-11E	Cenomanian	-28.4	1.9
AR1-11F	Cenomanian	-28.4	1.4
AR1-12A	Early Turonian	-25.0	2.1
AR1-14	Turonian	-27.9	1.6
AR1-17A	Coniacian	-28.0	4.6
AR1-17B	Coniacian	-27.8	4.0
AR1-20	Late Santonian	-26.6	1.0
AR1-21	Santonian?	-25.4	0.7
AR1-23	Late Campanian	-26.3	0.4
AR1-24	Paleocene	-25.9	1.6

•

 Table 2: Organic Matter Contents and Stable Carbon Isotope Values (TOC and δ13CTOC) of SedimentaryRocks Analysed in this Study
Sample		AR1-1	AR1-6	AR1-6 AR1-7 AR1-		AR1-9	AR1-10	AR1-11B	AR1-11C	AR1-11D
ľ	Age	Early Cretaceous	Late Albian	Late Albian	Cenomanian	Cenomanian	Cenomanian	Cenomanian	Cenomanian	Cenomanian
wt%										
SiO ₂		57.09	52.54	64.90	55.92	55.48	56.28	53.10	56.19	56.59
Al ₂ O ₃		19.72	19.43	15.71	19.78	17.17	11.80	19.78	19.54	17.54
Fe ₂ O ₃		6.60	7.25	4.40	6.05	7.06	7.92	6.43	5.44	5.94
MnO		0.04	0.02	< D.L.	0.02	0.02	0.04	0.02	0.02	0.02
MgO		2.23	2.28	2.06	1.72	2.34	1.11	2.18	1.74	1.91
	CaO	1.12	0.52	0.73	0.25	0.74	0.90	0.24	0.47	0.35
	Na ₂ O	2.30	1.45	3.55	1.07	1.45	0.49	1.42	1.72	2.01
I	K₂O	2.27	2.42	2.61	3.14	2.70	1.03	2.96	2.62	2.89
I	TiO ₂	0.93	0.94	0.70	1.06	0.88	0.69	1.28	0.95	1.27
I	P_2O_5	0.17	0.31	0.15	0.13	0.58	0.34	0.13	0.46	0.10
	LOI	7.67	12.71	5.46	10.91	11.46	16.75	12.79	11.28	10.76
Total		100.13	99.86	100.27	100.04	99.86	97.34	100.33	100.42	99.37
	Al/Si	0.39	0.42	0.27	0.4	0.35	0.24	0.42	0.39	0.35
	CIA	70	77	61	78	72	77	77	75	71
	ppm									
I	Sc	19.99	15.09	8.99	18.03	15.86	10.32	14.11	14.38	11.36
	Rb	100.58	122.28	84.48	152.45	125.62	61.58	155.82	125.04	142.04
	Cs	7.49	6.53	2.61	10.10	6.27	3.56	7.58	5.77	6.57
	Ва	1836.12	1277.34	2383.46	666.73	2829.03	26793.14	342.00	341.76	394.32
	Sr	206.01	109.76	195.54	103.58	123.21	269.75	82.50	100.41	106.65
	Th	5.45	8.86	5.02	10.01	9.21	6.74	10.36	11.67	9.20
	U	1.83	7.15	2.01	3.40	2.77	3.34	4.78	3.96	7.11
	Y	14.2	11.0	9.0	16.5	14.1	10.8	8.0	8.8	7.4
	Zr	113.51	125.81	111.17	180.52	135.19	134.04	155.75	172.29	208.16
	Nb	8.31	9.68	7.19	11.07	9.59	10.24	13.45	9.91	13.72
	Hf	3.08	3.65	3.04	5.06	3.90	3.53	4.59	4.88	5.96
	Cr	188.87	241.18	129.11	237.96	189.08	223.43	318.57	222.02	306.20
	v	131.11	170.71	96.99	149.10	189.91	224.00	328.04	253.35	261.33
	Co	17.68	12.74	8.74	16.64	9.34	8.26	19.35	9.87	15.84
	Cu	59.98	26.15	11.49	30.82	22.90	42.30	30.89	25.37	35.73
	Ni	83.74	58.08	37.68	50.53	50.53	41.69	84.40	53.40	83.96
	Zn	75.66	97.99	53.24	54.71	103.01	89.60	92.53	142.13	114.22
	La	21.07	18.82	14.63	36.97	22.94	17.86	13.87	13.48	10.28
	Ce	40.70	36.29	28.68	70.13	45.19	36.16	22.10	28.02	15.76
	Pr	4.53	4.41	3.31	8.08	5.43	4.50	2.55	3.53	1.77
	Nd	16.46	16.49	12.62	29.99	20.68	16.94	9.08	13.69	6.25
	Sm	2.95	3.27	2.46	5.20	3.96	3.32	1.69	2.69	1.26
I	Eu	0.77	0.80	0.73	1.24	1.01	1.21	0.41	0.61	0.33
	Gd	2.30	2.43	1.94	3.76	3.07	2.60	1.30	1.91	1.07
	Tb	0.38	0.35	0.29	0.53	0.46	0.37	0.21	0.28	0.18
	Dy	2.50	2.09	1.72	3.16	2.67	2.12	1.43	1.72	1.30
	Ho	0.55	0.44	0.35	0.63	0.53	0.41	0.33	0.36	0.32
	Er	1.60	1.21	0.99	1.76	1.46	1.07	1.02	1.03	1.01
	Im	0.25	0.18	0.15	0.26	0.21	0.15	0.17	0.17	0.17
	Yb	1.63	1.29	1.02	1.80	1.47	1.02	1.25	1.22	1.30
	Lu –	0.26	0.21	0.16	0.28	0.22	0.15	0.21	0.20	0.21
	la	0.88	0.96	2.32	1.52	0.98	0.94	1.31	0.97	1.34
	Sm/Nd	0.19	0.20	0.10	0.17	0.10	0.20	0.10	0.20	0.20
I	5/1/NU Eu/Eu*	0.10	0.20	1.02	0.17	0.19	1.20	0.19	0.20	0.20
I		1.00	0.07	0.00	0.00	0.09	0.07	0.04	0.02	0.00
I	1208 Cr/Th	34.6	0.90	0.99	0.90	20.5	0.97	0.09 30 g	10.90	0.00
I		0.27	0.50	0.56	23.0	0.59	0.65	0.72	0.91	0.00
I	7r/9c	5.69	Q 21	12 27	10.00	8 52	12 00	11 0/	11 09	18 22
I	IDEE	1.58	1 51	1 15	2.82	1.96	15	0.04	1 12	0.66
I	HREE	1.30	1 30	1 11	1 05	1.00	1.5	1 36	1 32	1 38
I	MRFF	1 55	1 45	1 18	22	1.85	1.52	0.88	1 16	0.76
I	MREE*	1.69	1.45	1 13	2 38	1 72	13	1 15	1.10	1 02
1		1.03	1.40	1.10	2.00	1.14	1.0	1.10	1.20	1.02

Table 3: Major and Trace Element Concentrations of Sedimentary Rocks Analyzed in this Study . D.L. : Detection Limit

Note. Uncertainties for all elements are given in the supplementary material (Table S2)

 $LREE = La_{PAAS} + Pr_{PAAS} + Nd_{PAAS} (in ppm); MREE = Gd_{PAAS} + Tb_{PAAS} + Dy_{PAAS} (in ppm);$

HREE = TmPAAS + YbPAAS + LuPAAS (in ppm); MREE* = (HREE+LREE) / 2;

 $CIA=[Al_2O_3/(Al_2O_3+CaO^*+Na_2O+K_2O)] \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu_N/(Sm_N \times Gd_N)^{1/2} \times 100 \text{ (in molar proportions); } Eu/Eu^* = Eu/Eu^*$

where XN refers to the chondrite-normalized concentration value [Condie, 1993];

 $\Omega Ce = 2 \ x \ (Ce_{sample}/Ce_{PAAS}) \ / \ [(La_{sample}/La_{PAAS}) + (Pr_{sample}/Pr_{PAAS})]$

Sample	AR1-11E	AR1-11F	AR1-12A	AR1-14	AR1-17A	AR1-17B	AR1-20	AR1-21	AR1-23	AR1-24
Age	Cenomanian	Cenomaniar	Early Turonian	Turonian	Coniacian	Coniacian	Early Santoniar	Santonian?	Late Campaniar	Paleocene
wt%										
SiO ₂	52.29	53.10	49.47	61.86	57.11	61.77	52.75	51.21	53.19	50.80
Al ₂ O ₃	20.00	19.04	22.95	15.43	13.45	13.13	20.50	21.98	20.75	14.83
Fe ₂ O ₃	6.10	6.01	5.35	4.91	3.98	4.16	7.57	7.83	6.96	8.09
MnO	0.02	0.02	< D.L.	< D.L.	< D.L.	< D.L.	0.03	0.03	0.02	0.02
MgO	2.30	1.69	1.76	1.78	0.91	1.00	1.89	1.92	1.80	1.07
CaO	0.35	0.95	0.19	0.29	1.54	0.68	0.26	0.26	0.31	2.60
Na₂O	1.43	1.34	0.55	0.40	0.43	0.31	0.79	0.97	0.81	0.22
K₂O	2.98	2.68	1.62	1.36	1.17	1.05	2.36	2.95	3.19	1.50
TiO ₂	1.34	1.07	0.95	0.65	0.50	0.47	1.02	1.08	0.95	0.68
P_2O_5	0.47	1.47	0.21	0.19	1.60	1.15	0.20	0.17	0.28	0.17
LOI	12.47	12.24	16.19	12.36	18.58	16.90	12.45	11.76	12.00	19.90
Total	99.75	99.60	99.22	99.23	99.26	100.62	99.80	100.16	100.26	99.87
Al/Si	0.43	0.41	0.53	0.28	0.27	0.24	0.44	0.49	0.44	0.33
CIA	76	74	89	85	74	82	83	81	80	69
ppm										
Sc	13.43	14.62	14.52	12.08	13.44	12.75	21.22	21.86	19.65	12.81
Rb	153.68	132.56	115.22	102.31	73.22	67.93	149.33	183.20	159.01	63.81
Cs	7.69	6.65	7.30	5.81	4.16	4.17	7.25	8.07	6.57	4.31
Ba	340.37	308.91	946.30	664.12	446.69	369.73	877.89	810.99	415.59	410.19
Sr	87.07	101.91	61.06	57.60	113.04	72.26	83.53	84.92	76.10	58.28
Th	14.39	16.37	9.87	7.68	9.84	9.72	17.25	21.49	19.67	13.35
U	5.49	12.01	4.35	3.30	15.54	9.38	3.97	4.44	4.86	4.27
Y	8.3	13.0	10.0	8.0	39.3	23.5	25.8	32.0	26.6	24.6
Zr	177.27	174.12	107.77	72.74	92.63	75.43	129.46	151.13	183.69	133.55
Nb	13.81	12.86	10.98	8.21	7.30	6.43	17.58	21.42	19.01	13.49
Hf	5.27	4.86	3.30	2.28	2.70	2.20	4.09	4.73	5.67	3.46
Cr	319.80	228.02	230.77	195.32	372.85	354.00	175.32	122.98	150.74	82.26
V	263.30	220.20	645.18	421.17	210.40	199.49	144.34	127.21	122.75	76.96
Co	14.68	11.18	10.03	8.88	5.16	5.12	12.10	14.05	11.89	19.01
Cu	31.50	25.16	20.33	18.32	36.12	26.80	26.05	25.31	21.44	27.25
Ni	75.29	53.16	56.40	47.64	73.86	60.67	37.91	35.60	31.67	49.59
Zn	106.40	80.21	68.22	67.48	114.85	98.88	79.63	74.47	69.55	53.10
La	12.36	22.79	19.02	16.84	38.51	26.80	54.96	68.63	54.75	47.31
Ce	21.00	49.30	32.55	31.10	01.50	40.51	107.93	150.54	109.94	90.33
Pr	2.59	0.20	3.04	3.00	1.01	5.75	12.22	15.70	12.30	11.14
Na	9.00	23.09	13.07	13.03	29.00	21.40	44.25	57.55	45.55	40.74
Sm E	0.46	4.70	2.47	2.30	0.79 1.20	4.00	1.00	10.39	0.10	1.50
Eu	1 / 9	3.35	1.97	1 75	5.11	3.32	6.00	7.06	6.06	5.87
Gu Th	0.23	0.47	0.27	0.24	0.75	0.40	0.00	1 14	0.00	0.86
	1/0	2.60	1.74	1 /7	4 70	3.00	5.07	6.54	5 14	4.91
Но	0.34	0.53	0.38	0.29	1.03	0.64	1.01	1 29	1.04	0.97
Er	1 04	1 40	1 10	0.20	2.80	1.76	2 74	3.41	2.83	2 49
Tm	0.17	0.20	0.17	0.13	0.38	0.24	0.39	0.48	0.41	0.35
Yh	1.25	1 42	1 23	0.90	2 46	1.57	2 65	3 26	2 79	2 31
1.0	0.19	0.22	0.19	0.14	0.36	0.23	0.39	0.48	0.42	0.33
Ta	1 34	1 22	1.06	0.77	0.69	0.61	1.63	2 03	1.84	1.34
14				0	0.00	0.01		2.00		
Sm/Nd	0.19	0.20	0.18	0.18	0.20	0.19	0.18	0.18	0.18	0.18
Eu/Eu*	0.83	0.82	0.83	0.78	0.72	0.72	0.68	0.67	0.68	0.71
ΩCe	0.92	0.99	0.92	0.96	0.85	0.90	1.00	1.01	1.01	1.01
Cr/Th	22.2	13.9	23.4	25.4	37.9	36.4	10.2	5.7	7.7	6.2
Th/Sc	1.07	1.12	0.68	0.64	0.73	0.76	0.81	0.98	1	1.04
Zr/Sc	13.2	11.91	7.42	6.02	6.89	5.92	6.1	6.91	9.35	10.43
LREE	0.92	2.05	1.36	1.26	2.82	2.02	4.2	5.38	4.28	3.77
HREE	1.32	1.52	1.3	0.96	2.66	1.71	2.83	3.48	2.99	2.47
MREE	0.95	1.93	1.15	1.02	3.13	2.03	3.56	4.66	3.6	3.48
MREE*	1.12	1.78	1.33	1.11	2.74	1.87	3.51	4.43	3.63	3.12

Table 2 (Continued)

Sample	Age	Sr (ppm)	⁸⁷ Sr/ ⁸⁶ Sr	± 1σ (*10 ⁶)	Nd (ppm)	¹⁴³ Nd/ ¹⁴⁴ Nd	±1σ	٤ _{Nd(0)}	±1σ	T _{DM} (Ga)
AR1-1	Early Cretaceous	206.01	0.726863	0.000005	16.46	0.512523	0.000016	-2.2	0.3	0.9
AR1-6	Late Albian	109.76	0.730869	0.000005	16.49	0.511549	0.0000047	-21.2	0.1	2.6
AR1-7	Late Albian	195.54	0.721161	0.000003	12.62	0.51167	0.000008	-18.9	0.2	2.3
AR1-8	Cenomanian	103.58	0.732824	0.000003	29.99	0.511674	0.000008	-18.8	0.2	2.1
AR1-9	Cenomanian	123.21	0.730814	0.000005	20.68	0.512608	0.000011	-0.6	0.2	0.8
AR1-10	Cenomanian	269.75	0.714598	0.000005	16.94	0.511694	0.000004	-18.4	0.1	2.3
AR1-11B	Cenomanian	82.50	0.746371	0.000006	9.08	0.511599	0.000005	-20.3	0.1	2.3
AR1-11C	Cenomanian	100.41	0.746923	0.000005	13.69	0.511581	0.000006	-20.6	0.1	2.5
AR1-11D	Cenomanian	106.65	0.722602	0.000005	6.25	0.511614	0.000009	-20.0	0.2	2.5
AR1-11E	Cenomanian	87.07	0.738315	0.000005	9.85	0.512015	0.000025	-12.2	0.5	1.8
AR1-11F	Cenomanian	101.91	0.738174	0.000004	23.89	0.511669	0.000006	-18.9	0.1	2.4
AR1-12A	Early Turonian	61.06	0.760897	0.000005	13.67	0.51178	0.00001	-16.7	0.2	2.0
AR1-14	Turonian	57.60	0.736446	0.000005	13.03	0.511634	0.000083	-19.6	1.6	2.2
AR1-17A	Coniacian	113.04	0.725604	0.000005	29.58	0.5117585	0.000011	-17.2	0.2	2.2
AR1-17B	Coniacian	72.26	0.730986	0.000005	21.46	0.511827	0.000026	-15.8	0.5	2.0
AR1-20	Late Santonian	83.53	0.753144	0.000004	44.25	0.511588	0.000006	-20.5	0.1	2.2
AR1-21	Santonian ?	84.92	0.773011	0.000006	57.53	0.511939	0.000006	-13.6	0.1	1.8
AR1-23	Late Campanian	76.10	0.774847	0.000006	45.55	0.512535	0.00008	-2.0	0.2	0.9
AR1-24	Paleocene	58.275	0.749076	0.000005	40.74	0.51159	0.000003	-20.4	0.1	2.3

Table 4: Nd–Sr isotopic systematics of sedimentary rocks analyzed in this study