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Marion Garçon, Catherine Chauvel, Christian France-Lanord, Mara Limonta, Eduardo Garzanti. Which minerals control the Nd–Hf–Sr–Pb isotopic compositions of river sediments?. *Chemical Geology*, 2014, 364, pp.42 - 55. 10.1016/j.chemgeo.2013.11.018 . hal-01769091

**HAL Id: hal-01769091**

**<https://hal.univ-lorraine.fr/hal-01769091>**

Submitted on 23 Aug 2022

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# Which minerals control the Nd-Hf-Sr-Pb isotopic compositions of river sediments?

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## 27 **Abstract**

28 River sediments naturally sample and average large areas of eroded continental  
29 crust. They are ideal targets not only for provenance studies based on isotopic  
30 compositions, but also can be used to establish average continental crust isotopic values.  
31 In large fluvial systems, however, mineral sorting processes significantly modify the  
32 mineralogy, and thus the geochemistry of the transported sediments. We still do not  
33 know, in any quantitative way, to what extent mineral sorting affects and fractionates the  
34 isotopic compositions of river sediments. Here, we focus on this issue and try to decipher  
35 the role of each mineral species in the bulk isotopic compositions of bedloads and  
36 suspended loads sampled at the outflow of the Ganga River that drains the Himalayan  
37 mountain range.

38 We analyzed Nd, Hf, Sr, and Pb isotopic compositions as well as trace element  
39 contents of a large number of pure mineral fractions (K-feldspar, plagioclase, muscovite,  
40 biotite, magnetite, zircon, titanite, apatite, monazite/allanite, amphibole, epidote, garnet,  
41 carbonate and clay) separated from bedload and bank sediments. We combine these data  
42 with mineral proportions typical of the Ganga sediments to perform Monte-Carlo  
43 simulations that quantify the contributions of individual mineral species to the Nd, Hf, Sr,  
44 and Pb isotopic budgets of bedloads and suspended loads.

45 We show that the isotopic systematics of river sediments are buffered by very few  
46 minerals. Despite their extremely low proportions in sediments, zircon and  
47 monazite/allanite control Hf and Nd isotopes, respectively. Feldspars, epidote, and  
48 carbonate buffer the Sr isotopic budget while clay, feldspars, and heavy minerals  
49 dominate Pb isotopes. Hafnium, Sr, and Pb isotopic differences between bedloads and  
50 suspended loads are well explained by their different mineral compositions. This

51 confirms that Hf, Sr and Pb isotopic compositions of sediments are strongly biased by  
52 mineral sorting processes during fluvial transport; hence they do not always constitute  
53 good proxies for provenance studies. In addition, we anticipate that fractionation of the  
54 isotopic systems continues at the river/ocean interface to deliver sediments to the deep  
55 ocean that are not necessarily similar to their crustal precursors, creating a systematic  
56 bias between the compositions of crustal sources and oceanic sediments.

57

58 **Keywords:** isotopic fractionation, bedload, suspended load, continental crust,  
59 Himalayas, minerals

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## 73 **1. Introduction**

74           Because they integrate large portions of continental crust exposed to weathering,  
75 river sediments from worldwide fluvial systems have been widely used to trace sediment  
76 provenance, estimate the average isotopic composition of the upper continental crust, and  
77 constrain its evolution through time (Goldstein et al., 1984; Goldstein and Jacobsen, 1988;  
78 Asmerom and Jacobsen, 1993; Allègre et al., 1996; Clift et al., 2002; Singh and France-  
79 Lanord, 2002; Millot et al., 2004; Kamber et al., 2005; Richards et al., 2005; Roddaz et al.,  
80 2005; Singh et al., 2008; Cina et al., 2009; Belousova et al., 2010; Hawkesworth et al., 2010;  
81 Wu et al., 2010; Dhuime et al., 2011; Padoan et al., 2011). Most studies assume that the  
82 isotopic compositions of river sediments reflect that of the drained continental area.  
83 However, depending on its size, shape, and density, detrital material transported by rivers  
84 is segregated within the water column by hydrodynamic processes: fast-settling coarse  
85 and heavy minerals concentrate in bedloads while fine, platy and light minerals are  
86 preferentially transported in suspension, and at different depths in the water column  
87 depending on their settling velocities (Singh and France-Lanord, 2002; Komar, 2007; Galy  
88 et al., 2008; Garzanti et al., 2008). Because chemical elements are carried in different  
89 amounts by the various minerals, it is now well described that mineral sorting processes  
90 lead to large chemical variability between bedload and suspended load sediments  
91 (Garzanti et al., 2010; Bouchez et al., 2011; Garzanti et al., 2011; Lupker et al., 2011; 2012).  
92 In contrast, our knowledge of how such processes affect the Nd, Hf, Sr and Pb isotopic  
93 compositions of sediments is relatively limited. While several studies have pointed out  
94 Nd, Hf, Sr and Pb isotopic fractionations between fine and coarse-grained sediments due  
95 to mineral sorting processes (Patchett et al., 1984; McLennan et al., 1989; Revel et al.,  
96 1996; Eisenhauer et al., 1999; Singh and France-Lanord, 2002; Chauvel et al., 2008; Bayon

97 et al., 2009; Carpentier et al., 2009; Chauvel et al., 2009; Vervoort et al., 2011; Garçon et  
98 al., 2013a, 2013b), the influence of each mineral species on the isotopic composition of  
99 sediments has never been thoroughly quantified.

100 In this paper, we report trace element contents and Nd, Hf, Sr and Pb isotopic  
101 compositions for a large number of pure mineral fractions separated from a bedload and  
102 a bank sediment sampled at the outflow of the Ganga fluvial system in the Bangladesh  
103 delta. Using Monte-Carlo simulations, we combine these data with mineral proportions  
104 reported by Garzanti et al. (2010; 2011) and Lupker et al. (2012) in river sediments  
105 sampled at the same location to (1) evaluate the individual contribution of each mineral  
106 species to the Nd, Hf, Sr and Pb isotopic budgets of bedloads and suspended loads and (2)  
107 determine if the known mineralogy of bedloads and suspended loads accounts for the  
108 observed difference in isotopic compositions. Finally, we discuss the implications of our  
109 results for sediment provenance studies based on isotope data and for large-scale isotopic  
110 partitioning between continental and oceanic sediments.

## 111 **2. Studied Area and Samples**

112 The studied mineral fractions were separated from two samples (bank sediment  
113 BGP 6 and bedload BR 717) collected at the outflow of the Ganga River, before the  
114 confluence with the Brahmaputra River (Figure 1). The sampling site was selected  
115 because it is located downstream, far away from the mountain range. As a consequence,  
116 the sediments integrate all materials transported by the Ganga River and its tributaries.  
117 These rivers belong to one of the largest fluvial systems on Earth (Milliman and Meade,  
118 1983), stretch over more than 2500 km and deliver about four hundred million tons of  
119 sediments to the ocean each year (Lupker et al., 2011). Such sediments are dominantly  
120 (>95%) derived from the Himalayan mountain range with minor contributions from the

121 Deccan Trapps and Indian craton supplied by the southern tributaries of the Ganga (Galy  
122 and France-Lanord, 2001; Singh et al., 2008; Lupker et al., 2011). Himalayan materials  
123 mainly consist of sedimentary series and crystalline metamorphic rocks (e.g., Le Fort,  
124 1975). Because of the large lithological and chemical diversity of Himalayan rocks, the  
125 Ganga basin can be considered as a representative portion of the upper continental crust.  
126 All minerals analyzed in our study are thus ubiquitous in the erosion products of any  
127 upper continental crust exposed to weathering (McLennan, 1989; Taylor and McLennan,  
128 1995; McLennan, 2001) and our results should be relevant for river sediments sampled  
129 in other worldwide fluvial systems draining large continental areas.

130         The second major advantage of the selected site is that these sediments were  
131 extensively studied for their mineralogy by Garzanti et al. (2010; 2011) and Lupker et al.  
132 (2012) (Supplementary Table 1). These authors showed that bedloads and suspended  
133 loads share quite similar types of minerals but in different proportions. Surface  
134 suspended loads are generally finer and relatively rich in micas and clays while bedloads  
135 tend to be coarser and richer in quartz and heavy minerals. Among the heavy minerals  
136 that are commonly found in the Ganga sediments, amphibole, epidote and garnet are the  
137 most abundant but many others are also present: titanite, zircon, rutile, apatite, monazite,  
138 tourmaline and allanite for example (Garzanti et al., 2010; 2011). Detailed mineral  
139 proportions in the Ganga bedloads and suspended loads are listed in Supplementary  
140 Table 1.

141         Finally, several studies have reported major and trace element contents, as well as  
142 Nd, Hf, and Pb isotopic compositions, of bulk suspended loads and bedloads from this  
143 sampling site (Galy and France-Lanord, 2001; Lupker et al., 2011; 2012; Garçon et al.  
144 2012, 2013a; 2013b; see Supplementary Table 2). We used these data to evaluate the role  
145 of individual minerals in the bulk isotopic budget of river sediments. Because no Sr

146 isotopic data exist in the literature for unleached bedloads and suspended loads, we  
147 analyzed the bulk Sr isotopic compositions of two bedloads (BR 717 and BR 418), one  
148 bank sediment (BGP 6) and 2 suspended loads (BR 414 and BR 412) sampled in the  
149 Bangladesh delta (Figure 1). The trace element data and the Nd, Hf, and Pb isotopic  
150 compositions of these five samples are reported in Supplementary Table 2. The detailed  
151 sampling methods and the major element data can be found in Galy and France-Lanord  
152 (2001), Lupker et al. (2011; 2012) and Garçon et al. (2013a; 2013b). We also analyzed the  
153 Sr isotopic compositions of several grain-size fractions separated from a suspended load  
154 from the Ganga River (sample BR 522, Figure 1) to help us interpreting the Sr results of  
155 the Monte-Carlo simulations.

### 156 **3. Methods**

#### 157 **3.1. Mineral separation**

158 The starting material consists of several kilos of sediment collected during the  
159 monsoon season. With the exception of the clay fraction separated from bank sediment  
160 BGP 6, all mineral fractions were separated from bedload BR 717 dredged in the main  
161 channel of the Ganga River. This sample was first washed and dried, then divided into 3  
162 grain-size fractions by sieving: 63-100  $\mu\text{m}$ , 125-250  $\mu\text{m}$ , and >250  $\mu\text{m}$ . For each fraction,  
163 we used successive centrifugations in heavy liquids (sodium metatungstate and  
164 methylene iodide) to separate minerals and partial freezing in liquid nitrogen to recover  
165 them. After this step, some of the minerals were isolated using a hand magnet or the  
166 Frantz isodynamic magnetic separator. Biotite, muscovite, plagioclase, quartz, and K-  
167 feldspar were isolated from the >250  $\mu\text{m}$  fraction; magnetite, amphibole, apatite, garnet,  
168 epidote, titanite, monazite, allanite, and zircon from the 63-100  $\mu\text{m}$  and/or the 125-250



169  $\mu\text{m}$  fractions. Each mineral fraction was finally carefully purified under the binocular and  
170 polarizing microscopes to remove impurities.

171 Because monazite and allanite were very small and difficult to distinguish from  
172 each other, we analyzed an impure fraction containing high amounts of both minerals. In  
173 the rest of the paper, this fraction is called the “monazite/allanite fraction”. Similarly, we  
174 could not remove all impurities in the apatite fraction; hence we analyzed an impure  
175 fraction highly enriched in apatite. Following Galy et al. (1999), the composition of the  
176 carbonate fraction was obtained using a 1N acetic acid leaching procedure for one hour  
177 in an ultrasonic bath. To ensure a pure carbonate fraction and avoid dissolution of other  
178 minerals or contamination by clay minerals, we leached a coarse fraction containing only  
179 carbonate, quartz, K-feldspar, plagioclase, and few phyllosilicates (non-magnetic 125-250  
180  $\mu\text{m}$  fraction with a density  $< 2.90 \text{ g.cm}^{-3}$ ). The carbonate-rich leachate was then isolated  
181 by centrifugation and evaporated on a hot plate. Finally, a clay fraction, selected as the  $< 2$   
182  $\mu\text{m}$  fraction, was separated by sieving and settling from bank sediment BGP 6 because we  
183 lacked sufficient material to recover it from bedload BR 717.

184 For each mineral species, only one fraction was separated and analyzed; the only  
185 exception is the K-feldspar fraction (Table 1). We recognized that multiple analyzes of the  
186 same mineral species could have been interesting to assess the compositional  
187 heterogeneity of the Himalayan minerals. However, due to the limited quantity of starting  
188 material and the time-consuming work involved in the handpicking procedure we could  
189 not perform other duplicate analyses. We are however confident that the measured  
190 fractions should be representative of the mineral populations because for each mineral  
191 separate, the number of grains present in the analyzed fraction was always higher than a  
192 thousand. For example, the 5mg fraction of zircons analyzed in this study most probably  
193 contains between 1,000 to 2,000 zircon grains assuming that grains are spherical with a

194 mean diameter of 50-60  $\mu\text{m}$ , as suggested by Garzanti et al. (2010; 2011) and Garçon et  
195 al. (2013b).

196

### 197 **3.2. Chemistry**

198 For each mineral separate, we performed a single dissolution used to analyze both  
199 trace element concentrations and Nd, Hf and Sr isotopes. Note that Pb isotopic  
200 compositions of these sediments were previously published by Garçon et al. (2013b).  
201 Methods used to obtain Pb data are described in this latter paper.

202 After washing with ultrapure water, 5 to 100 mg of mineral grains or powder were  
203 first attacked for 2 days in 14N  $\text{HNO}_3$  on a hot plate at 130°C, then evaporated and  
204 dissolved in a mixture of HF and  $\text{HClO}_4$  in teflon containers placed in steel PARR bombs  
205 for 10 days at 200°C to ensure complete dissolution. For bulk bedloads and suspended  
206 loads, 50 to 100 mg of fine powder were dissolved using the same procedure as for the  
207 mineral fractions. Finally, the bulk suspended load BR 522 and its grain-size fractions,  
208 separated using settling and sieving techniques, were leached in 1N acetic acid for one  
209 hour in an ultrasonic bath to remove the carbonates before being digested following the  
210 same dissolution procedure as the one described above.

211 We measured trace element concentrations by ICP-MS with a dilution factor of  
212 5000 and the equivalent of 2mg of sample. Measurements were performed using an  
213 Agilent 7500ce ICP-MS in Grenoble (France). Oxide interference and analytical drift were  
214 corrected following the procedure of Chauvel et al. (2011). BR-24 rock standards were  
215 measured every four samples to calibrate and convert peak signals into concentrations  
216 offline. To ensure the validity of our results, international rock standards were run as

217 unknown samples, including sedimentary materials such as JSd-2 (see Supplementary  
218 Table 3).

219 Hafnium, Nd and Sr were isolated from the same aliquot using the ion resin  
220 techniques and following the procedure described by Chauvel et al. (2011). Hafnium was  
221 recovered in three steps using a first column filled with cationic resin (AG50W-X8) to  
222 isolate the High Field Strength Elements (HFSE) followed by a second anion resin column  
223 (AG1-X8) and a third cation resin column (AG50W-X8) to isolate Hf from the other HFSE.  
224 Nd was recovered in two steps: a first cation resin column (AG50W-X8) to isolate the Rare  
225 Earth Elements (REE) and a second column filled with Ln-Spec resin to separate Nd from  
226 the other REE. Finally, Sr was isolated using two successive columns filled with cationic  
227 resin (AG50W-X8) and Sr-Spec resin. Total procedural blanks were < 80 pg for Nd (n=1),  
228 < 35 pg for Hf (n=2) and < 140 pg for Sr (n=2). These small amounts can be considered  
229 negligible compared to the amount of Nd, Hf and Sr isolated in all samples. Nd and Hf  
230 isotopes were measured on a Nu Plasma MC-ICP-MS at the ENS Lyon (France). Nd isotopic  
231 compositions were normalized to  $^{146}\text{Nd}/^{144}\text{Nd}=0.7219$  and Hf isotopic compositions to  
232  $^{179}\text{Hf}/^{177}\text{Hf}=0.7325$ . The Ames-Rennes Nd and Ames-Grenoble Hf reference standards run  
233 every two or three samples yielded average  $^{143}\text{Nd}/^{144}\text{Nd} = 0.511953 \pm 8$  ( $2\sigma$ ,  $n = 15$ ) and  
234  $^{176}\text{Hf}/^{177}\text{Hf} = 0.282157 \pm 18$  ( $2\sigma$ ,  $n = 14$ ), respectively. We used the recommended values  
235 published by Chauvel and Blichert-Toft (2001) and Chauvel et al. (2011) for these two  
236 standards to correct the analytical drift all through the measurement sequence. Sr  
237 isotopic compositions were measured using a solid source ThermoScientific Triton mass  
238 spectrometer in Brest (France) and normalized to  $^{86}\text{Sr}/^{88}\text{Sr}=0.1194$ . The NBS 987  
239 reference material yielded average  $^{87}\text{Sr}/^{86}\text{Sr} = 0.710270 \pm 7$  ( $2\sigma$ ,  $n = 10$ ) and values  
240 measured for the samples were not normalized.

## 241 **4. Results**

### 242 **4.1. Trace element concentrations**

243 Trace element concentrations measured in the mineral fractions are reported in Table 1  
244 and shown in Figure 2 as spidergrams normalized to the average composition of upper  
245 continental crust (Rudnick and Gao, 2003). Except for mobile elements such as Cs, Rb, Ba,  
246 Sr, Li, Co, or Ni, heavy minerals (Figure 2a) generally display higher trace element  
247 contents than light minerals (Figure 2b). The trace element patterns of the various  
248 minerals are generally consistent with published values (e.g., Götze and Lewis, 1994; Bea,  
249 1996; Garçon et al., 2011; Garzanti et al., 2011). Zircon and garnet show typical depletions  
250 in light REE while monazite/allanite show depletion in heavy REE. The REE distributions  
251 of other minerals, including light minerals, are rather flat. The monazite/allanite and  
252 zircon separates clearly exhibit the highest concentrations in LREE (including Nd) and Hf,  
253 respectively, with enrichment factors > 1000 (see Figure 2). The highest Pb contents are  
254 found in clay (95 ppm) and K-feldspar (75 ppm). The highest Sr concentrations are found  
255 in epidote (1509 ppm), carbonate (526 ppm), K-feldspar (243 ppm) and plagioclase (195  
256 ppm). Both apatite and monazite/allanite fractions display lower REE and higher Zr, Hf  
257 concentrations compared to what could be expected in pure apatite and  
258 monazite/allanite separates (Ayres and Harris, 1997; Garçon et al., 2011; Garzanti et al.,  
259 2011), a feature probably due to the presence of impurities such as light minerals and  
260 zircon grains. This can easily be explained by the fact that we were unable to purify the  
261 apatite and monazite/allanite fractions under the binocular microscope due to the very  
262 small size of the minerals. Similarly, the quartz fraction contains anomalously high REE  
263 and Sr concentrations compared to what is usually obtained in pure quartz (see for

264 example Monecke et al., 2000 and Garzanti et al., 2010). It is thus likely that this fraction  
265 also contains impurities.

266

## 267 **4.2. Isotopic compositions**

268 The Nd, Hf and Sr isotopic compositions for all mineral separates are reported in  
269 Table 1. No Hf isotopic composition could be measured for the carbonate fraction because  
270 of its very low Hf concentration. We also did not measure the isotopic compositions of the  
271 quartz fraction and the Hf and Sr isotopes of the monazite/allanite-rich and the apatite-  
272 rich separate because of the presence of other minerals in these separates that could  
273 strongly influence the measured isotopic compositions.

274 Nd, Hf and Sr isotopic compositions span large ranges of values between 0.511732  
275 and 0.512235 for  $^{143}\text{Nd}/^{144}\text{Nd}$ , 0.281970 and 0.282803 for  $^{176}\text{Hf}/^{177}\text{Hf}$  and 0.7125 and  
276 0.8313 for  $^{87}\text{Sr}/^{86}\text{Sr}$ . The monazite/allanite fraction has the least radiogenic Nd isotopic  
277 compositions of all minerals while the zircon separate has the least radiogenic Hf isotopic  
278 compositions. Biotite and muscovite, known for their very high Rb/Sr ratios, have  
279 accordingly highly radiogenic Sr isotopic compositions (0.8123 and 0.8313, respectively).  
280 By contrast, epidote and carbonate have the least radiogenic Sr isotopic compositions  
281 (0.7125 and 0.7139) of all minerals.

282 Sr isotopic compositions measured in bulk bedloads, banks and suspended loads  
283 from the Ganga River are listed in Table 2. They are particularly radiogenic with  $^{87}\text{Sr}/^{86}\text{Sr}$   
284 varying between 0.7546 and 0.7737 and bedloads tending to have less radiogenic Sr  
285 isotopes than suspended loads. These results fall within the large range of Sr isotopes  
286 usually measured in the silicate and/or carbonate fractions isolated from Himalayan  
287 derived sediments (Harris et al., 1998; Galy et al., 1999; Galy and France-Lanord, 2001;  
288 Pierson-Wickmann et al., 2001; Singh et al., 2008). Such radiogenic isotopic compositions

289 are due to the presence of extremely radiogenic sources in the Himalayan mountain  
290 range, in particular in the Lesser Himalayan units ( $^{87}\text{Sr}/^{86}\text{Sr} > 0.8$ ; Derry and France-  
291 Lanord, 1996; Singh et al., 2008).

292 Sr isotopic compositions measured in the silicate fractions of the suspended load  
293 BR 522 and its grain-size fractions are provided in Table 2. The bulk leached suspended  
294 load BR 522 shares comparable  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios with the other unleached suspended loads  
295 from the Ganga River (Table 2). Significant Sr isotopic differences are however observed  
296 between the bulk sample and its grain-size fractions with the 2-20  $\mu\text{m}$  fraction being much  
297 more radiogenic than the others.

## 298 **5. Discussion**

299 River sediments are basically mixtures of mineral species. Some minerals, such as  
300 quartz, contain virtually no trace elements and their proportion in the sedimentary  
301 material influences only the silica content of the sediment but has no impact on most  
302 isotopic ratios. In contrast, other minerals very rich in certain trace elements or with very  
303 unusual isotopic compositions have the potential to significantly contribute or can even  
304 overwhelm the trace element and isotopic budget of river sediments even if they are  
305 present in very low proportions. A proper understanding of the role of individual mineral  
306 species in the isotopic budget of river sediments thus requires the consideration of  
307 several parameters. These include the nature and the proportion of the different mineral  
308 species as well as their trace element concentration and isotopic composition.

309 Here, we present Monte-Carlo simulations that quantitatively evaluate the role of  
310 individual minerals in the final Nd, Hf, Sr and Pb isotopic budget of river sediments. We  
311 simulate the composition of a hundred thousand mixtures between common mineral  
312 species that are present in the sedimentary material and that have the potential to

313 significantly influence its isotopic compositions. Then, we compare our results to the data  
314 published on bulk sediments.

315 Using mineral proportions estimated by Garzanti et al. (2010; 2011) and calcite  
316 contents measured by Lupker et al. (2012) in suspended loads and bedloads from the  
317 Ganga River in the Bangladesh delta, we first defined a range of possible proportions for  
318 each type of mineral (see Supplementary File). For each mineral species, we then  
319 randomly sample proportions within this range and associate them with the Nd, Hf, Sr or  
320 Pb concentrations and isotopic compositions measured in our mineral separates. Finally,  
321 we calculate the isotopic compositions of the resulting mixtures using the following  
322 equation, written here for the Nd isotopic system:

$$323 \quad \left( \frac{^{143}\text{Nd}}{^{144}\text{Nd}} \right)_{\text{mixture}} = \sum_{i=1}^n \left[ x_i \cdot \frac{C_{\text{Nd},i}}{C_{\text{Nd},\text{mixture}}} \cdot \left( \frac{^{143}\text{Nd}}{^{144}\text{Nd}} \right)_i \right]$$

324 where n is the number of minerals involved in the mixture;  $x_i$  the weight proportion of  
325 mineral i sampled randomly in a defined range;  $C_{\text{Nd},i}$  and  $\left( \frac{^{143}\text{Nd}}{^{144}\text{Nd}} \right)_i$  the Nd content  
326 and the Nd isotopic composition of mineral i (see Table 1); and  
327  $C_{\text{Nd},\text{mixture}} = \sum_{i=1}^n [x_i \cdot C_{\text{Nd},i}]$ , the Nd concentration of the mixture.

328 In the following sections, we compare the modeled Nd, Hf, Sr and Pb isotopic  
329 compositions to those measured in both suspended load and bedload sediments as  
330 sampled in the Ganga River in the Bangladesh delta (data from the literature and our Sr  
331 isotope measurements). Using such approach, we can evaluate the role of each mineral in  
332 the isotopic budget of all river sediments. We illustrate the results of our Monte-Carlo  
333 simulations for each isotopic system in diagrams that show the contribution of the various  
334 mineral species as a function of the modeled isotopic composition for all mixtures. The

335 contribution of each mineral  $i$  in the  $^{143}\text{Nd}/^{144}\text{Nd}$  budget, for example, is determined in  
336 percentage using the following equation:

$$337 \quad \text{Mineral contribution} = 100 \cdot x_i \cdot \frac{C_{\text{Nd},i}}{C_{\text{Nd,mixture}}} \cdot \left( \frac{^{143}\text{Nd}}{^{144}\text{Nd}} \right)_i \bigg/ \left( \frac{^{143}\text{Nd}}{^{144}\text{Nd}} \right)_{\text{mixture}}$$

338 All details about the input parameters, the way simulations were performed and  
339 the robustness of the Monte-Carlo results are provided in the Supplementary File. We  
340 performed several simulations to assess the sensitivity of our models to the proportions,  
341 concentrations and isotopic compositions of the minerals (see Supplementary File). Here,  
342 we just emphasize that the simulation results are very well constrained for the four  
343 isotopic systems. The contribution of each mineral species in the whole-rock isotopic  
344 budgets remains quite constant even if we introduce large errors on the input parameters,  
345 such as the mineral proportions or the concentrations and isotopic compositions of  
346 mineral species. An important implication is that the results presented in this study may  
347 be valid not only for the Ganga sediments, but also for any sediment sampled in other  
348 large river systems all over the world.

349

### 350 **5.1. Which minerals control Nd isotopes of river sediments?**

351 To perform the Monte-Carlo simulation modeling Nd isotopic compositions of  
352 sediments, we considered the 14 analyzed minerals (K-feldspar, plagioclase, muscovite,  
353 biotite, magnetite, zircon, titanite, apatite, monazite/allanite, amphibole, epidote, garnet,  
354 carbonate, and clay) and we set the mineral proportions within the range known for river  
355 sediments sampled in the Bangladesh delta (see Supplementary File and inset in Figure  
356 3). Results are shown in Figure 3 while  $^{143}\text{Nd}/^{144}\text{Nd}$  for the most important mineral  
357 phases are shown along the horizontal axis.



358           The first important observation is that minerals that significantly contribute to the  
359 Nd isotopic budget are very few: monazite/allanite, clay, titanite and biotite. All other  
360 minerals (K-feldspars, plagioclase, muscovite, magnetite, zircon, apatite, amphibole,  
361 epidote, garnet, and carbonate) always contribute to less than 10% of the isotopic budget,  
362 over the entire range of modeled Nd isotopic compositions; this is why they are not shown  
363 in Figure 3. Because of its extreme enrichment in Nd (Figure 2), the monazite/allanite  
364 fraction entirely dominates the Nd isotopic budget of modeled mixtures and it can even  
365 control 100% of the isotopic budget (Figure 3) while its proportion in the mineral  
366 assemblage never exceeds 0.5 wt.% (see Supplementary File and inset of Figure 3). It is  
367 only when the modeled Nd isotopic composition is greater than 0.51185 that the  
368 contribution of clay, titanite and biotite starts to surpass that of monazite and allanite  
369 (Figure 3).

370           As shown by the grey band in Figure 3, Nd isotopic ratios reported for bedloads  
371 and suspended loads sampled at the outflow of the Ganga span a limited range of values  
372 between 0.51172 and 0.51179 (Garçon et al., 2013a), values similar to those reported by  
373 Galy and France-Lanord (2001) for sediments sampled at the same location. According to  
374 our simulation (Figure 3), such compositions correspond to mixtures highly dominated  
375 by monazite and allanite (60-100%) with a small but significant contribution from clays  
376 (0-30%). We can therefore suggest that the few monazite and allanite grains present in  
377 the sediments (<0.5 wt%, see Supplementary File) totally overwhelm the Nd isotopic  
378 compositions of river sediments in the Bangladesh delta.

379           In a more classical diagram showing  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios as a function of  
380  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios (Figure 4), it becomes even more obvious that the unradiogenic Nd  
381 isotopic signature of the monazite/allanite fraction is the controlling factor of the  
382 composition of the Ganga sediments since no other mineral has a Nd isotopic composition

383 similar to or lower than that of the bulk sediments. As suggested by our Monte-carlo  
384 simulation, both bedloads and suspended loads are well reproduced by a mixture of  
385 monazite, allanite, clay  $\pm$  biotite, titanite (Figure 4).

386

## 387 **5.2. Which minerals control Hf isotopes of river sediments?**

388 The results of the Monte-Carlo simulation for Hf isotopes are shown in Figure 5  
389 together with the results obtained on the various mineral separates. Eleven minerals  
390 were introduced in the simulation: K-feldspar, plagioclase, muscovite, biotite, magnetite,  
391 zircon, titanite, amphibole, epidote, garnet, and clay. Quartz and carbonates were ignored  
392 because they do not contain any Hf and we did not measure their Hf isotopic  
393 compositions. The monazite/allanite fraction and the apatite fraction were also ignored  
394 because they contain a lot of small zircons, a feature that explains their unusually high Zr-  
395 Hf concentrations compared to what is commonly measured in such minerals (Ayres and  
396 Harris, 1997). We suggest, therefore, that if we had measured their Hf isotopic  
397 compositions, the results would not have been representative of the composition of the  
398 monazite, allanite or the apatite but would have reflected that of the zircons present in  
399 the fractions.

400 As for Nd isotopes, only few minerals significantly contribute to the Hf isotopic  
401 budget of the modeled mixtures: zircon, clay, muscovite, biotite and titanite. By contrast,  
402 garnet, amphibole, epidote, magnetite, plagioclase, and K-feldspar contribute to less than  
403 10% of the Hf isotopic budget and are not shown in Figure 5. Modeled Hf isotopic  
404 compositions span a relatively large range of values between 0.28197 and 0.28235. One  
405 of the most remarkable features is that unradiogenic Hf-rich zircon totally buffers the Hf  
406 isotopic budget over a large range of isotopic compositions (Figure 5) while its proportion

407 in the mineral assemblage is always less than 0.5 wt% (see Supplementary File and inset  
408 of Figure 5). Clay is the second important mineral but its contribution never exceeds 50%.  
409 Together with muscovite, biotite and titanite, the clay fraction accounts for the most  
410 radiogenic Hf isotopic compositions in the simulation.

411 In contrast to Nd, there is a difference between the  $^{176}\text{Hf}/^{177}\text{Hf}$  ratio of bedloads and  
412 suspended loads sampled at the outflow of the Ganga River (Garçon et al., 2012; 2013a).  
413 As shown by grey bands in Figure 5, bedloads are less radiogenic than suspended loads.  
414 The results of our Monte-carlo simulation show that the difference is easily explained by  
415 different contributions of unradiogenic zircon. The range of  $^{176}\text{Hf}/^{177}\text{Hf}$  measured in  
416 bedloads results from a very high contribution of zircon (i.e. 70-100% of their Hf isotopic  
417 budget) and the relative contribution of all other minerals remains always below 15%.  
418 The higher  $^{176}\text{Hf}/^{177}\text{Hf}$  measured in suspended loads results from a higher contribution  
419 of more radiogenic minerals such as clay, muscovite, biotite and titanite and a lower  
420 contribution of zircon, even if it can still contribute to up to 50% of the isotopic budget  
421 (Figure 5).

422 In Figure 6, we show the relationship between  $^{176}\text{Hf}/^{177}\text{Hf}$  and  $1/\text{Hf}$  for minerals,  
423 bedloads and suspended loads. In contrast to what we did for the Nd case, we chose to not  
424 represent Hf isotopic compositions as a function of their parent/daughter ratios (i.e.  
425  $^{176}\text{Lu}/^{177}\text{Hf}$  ratios) because Lu and Hf are not carried by the same minerals and it would  
426 introduce confusion when evaluating the role of individual minerals in the Hf isotopic  
427 compositions of sediments. We also normalized the  $1/\text{Hf}$  ratio of all sediments to the  $\text{SiO}_2$   
428 content of the clay fraction (see footnote of Figure 6 for more details), because dilution by  
429 variable amounts of quartz has a drastic effect on the Hf concentration of river sediments.  
430 As highlighted by our Monte-Carlo simulations, Figure 6 shows clearly that all bedloads  
431 plot close to zircon, confirming that this mineral buffers the Hf isotopic budget of

432 bedloads. The composition of suspended loads requires a mixture of more radiogenic  
433 minerals such as biotite, muscovite and clay together with less radiogenic titanite and  
434 zircon for some of them (Figure 6).

435

### 436 **5.3. Which minerals control Sr isotopes of river sediments?**

437 In Figure 7a, we show  $^{87}\text{Sr}/^{86}\text{Sr}$  measured on the various minerals and the modeled  
438  $^{87}\text{Sr}/^{86}\text{Sr}$  for mixing simulations between K-feldspar, plagioclase, muscovite, biotite,  
439 magnetite, zircon, titanite, amphibole, epidote, garnet, carbonate, and clay. Six minerals  
440 (garnet, amphibole, titanite, zircon, magnetite, and biotite) are not shown in the figure  
441 because they all are minor contributors (<10%) to the Sr isotopic budget. In contrast,  
442 because of their high Sr contents (Table 1), epidote, K-feldspar, plagioclase, carbonate  
443 and, to a lesser extent, clay, and muscovite exert a major role on the  $^{87}\text{Sr}/^{86}\text{Sr}$  of the  
444 modeled mixtures.

445 The range of Sr isotopic compositions measured in bedloads and suspended loads  
446 from the Ganga River is shown by grey bands in Figure 7a. Variations of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios  
447 as a function of  $1/\text{Sr}$  are reported in Figure 8. Figure 7a demonstrates clearly that our  
448 Monte-Carlo simulation fails in reproducing the high  $^{87}\text{Sr}/^{86}\text{Sr}$  of all sediments, in  
449 particular those of suspended loads. We can think of several possible reasons to explain  
450 the mismatch between modeled and measured Sr isotope ratios.

451 The first possibility is that we have missed a component/mineral with high  $^{87}\text{Sr}/^{86}\text{Sr}$   
452 and high Sr content in our Monte-Carlo procedure. The missing component cannot be  
453 dissolved Sr, or Sr carried by small particles (< 0.2  $\mu\text{m}$ ) because all suspended loads were  
454 filtered under pressure at 0.2  $\mu\text{m}$  (Lupker et al., 2011) and the filtered fractions  
455 containing particles smaller than 0.2  $\mu\text{m}$  were not analyzed in this study. As a

456 consequence, dissolved ions and very fine particles cannot participate to the Sr isotopic  
457 budget of suspended loads. Even if we assume that the filtration was not complete, the Sr  
458 isotopic composition of these fractions (clay < 2  $\mu$  m at 0.747 and river water with  
459  $^{87}\text{Sr}/^{86}\text{Sr} < 0.73$ , see Table 1 and Galy et al., 1999) are less radiogenic than the sediments,  
460 hence they cannot constitute the missing components with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. Given  
461 their high Sr contents (Table 1), potential other candidates are apatite and  
462 monazite/allanite. However, these minerals have low  $^{87}\text{Rb}/^{86}\text{Sr}$  ratios, just like carbonate  
463 and epidote that have the lowest  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (Figure 8). It seems thus unlikely that  
464 these phases could have the elevated  $^{87}\text{Sr}/^{86}\text{Sr}$  that is required to explain the observed  
465 mismatch between model and measurements. Adding these mineral phases in the  
466 simulations would probably even make the modeled Sr isotopic compositions less  
467 radiogenic.

468 A second possibility is that we underestimated the proportions of very radiogenic  
469 minerals, such as muscovite or biotite (Figure 8), in the fine suspended loads. However,  
470 as shown in details in the Supplementary File, the results of the Monte-Carlo simulations  
471 are robust and the range of modeled Sr isotopic compositions remains incompatible with  
472 the measured values even with very different mineral proportions.

473 The last possibility is that one or several mineral phases that contribute significantly  
474 to the Sr isotopic budget have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios higher than the value that we measured.  
475 We suspect that it could be the case for K-feldspar and muscovite for the following reason:  
476 for convenience, we separated these two minerals from the coarsest fraction (>250  $\mu$ m)  
477 of bedload BR 717 (see section 3.1) and we suspect that smaller minerals might have  
478 different isotopic compositions. Indeed, large ranges of  $^{87}\text{Sr}/^{86}\text{Sr}$  are reported for K-  
479 feldspar (0.737 to 0.801) and muscovite (0.765 to 8.353) in the Himalayan rocks (Mehta,  
480 1977; Kai, 1981; Ferrara et al., 1983; Deniel et al., 1987; Johnson and Rogers, 1997) and

481 we believe that the coarse feldspar and muscovite that we isolated ( $^{87}\text{Sr}/^{86}\text{Sr}$  of 0.7587  
482 and 0.8313, respectively) might have lower  $^{87}\text{Sr}/^{86}\text{Sr}$  than grains present in fine  
483 suspended loads.

484 Analyses of finer K-feldspar and muscovite fractions would help support this  
485 hypothesis but the separation and purification of a significant amount of fine minerals  
486 constitute a difficult technical challenge. As an alternative, we performed additional Sr  
487 isotopic analyses on several grain-size fractions separated from a Ganga suspended load  
488 sampled in the Bangladesh delta (sample BR 522; Figure 1, Table 2). These analyses were  
489 carried out on the silicate part of the samples to establish whether the Sr isotopic  
490 compositions of the silicate minerals could vary as a function of mineral grain-size.  
491 Results clearly indicate that the grain-size fractions do not have the same Sr isotopic  
492 composition as the bulk sample and span a relatively large range of values from 0.7575 to  
493 0.7824 (Table 2). Interestingly, the 2-20  $\mu\text{m}$  fraction, representing almost half of the  
494 suspended load sample (see Table 2), has the most radiogenic Sr isotopic composition,  
495 supporting the idea that finer K-feldspars and muscovites may be more radiogenic than  
496 the coarse minerals that we analyzed. This could occur if fine K-feldspars and muscovites  
497 were older than coarse ones. Calculations show that an age difference of only 50-70 Ma is  
498 sufficient to increase the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of muscovite from 0.8 to 1.0 assuming a  $^{87}\text{Rb}/^{86}\text{Sr}$   
499 ratio of 200-300, values commonly measured in Himalayan muscovite (e.g. Mehta, 1977;  
500 Kai, 1981; Ferrara et al., 1983; Deniel et al., 1987; Johnson and Rogers, 1997). The age  
501 difference required to increase the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of K-feldspar from 0.76 to 0.80 is of  
502 course higher (about 150 Ma) because  $^{87}\text{Rb}/^{86}\text{Sr}$  is usually around 20 (e.g. Ferrara et al.,  
503 1983; Deniel et al., 1987; Johnson and Rogers, 1997). Another possibility is that the fine  
504 K-feldspars and muscovites had initially crystallized with high Rb but similar Sr contents,  
505 leading to higher Rb/Sr ratios than the ratio measured in the coarse minerals, and leading

506 in turn, to more radiogenic Sr isotopic compositions over the same amount of time.  
507 Unfortunately, available data are not sufficient to constrain the origin of the observed  
508 isotopic variability as a function of grain-size and further work is needed to understand  
509 this feature.

510 In order to reproduce the isotopic compositions measured on whole-rock  
511 sediments, we revised the Monte-Carlo simulation assuming that K-feldspars and  
512 muscovites had more radiogenic Sr isotopic compositions but similar Sr concentrations.  
513 The results of the simulation using  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of 0.800 for K-feldspar (instead of  
514 0.7587) and 1.0 for muscovite (instead of 0.8313) are shown in Figure 7b. In this case, the  
515 simulated mixtures overlap with the fields known for bedloads and suspended loads. As  
516 in the first simulation (Figure 7a), K-feldspar, carbonate, clay, plagioclase, epidote and  
517 muscovite control the Sr isotopic budget of sediments (Figure 7b). Assuming that the  
518 revised simulation is valid, isotopic differences between bedloads and suspended loads  
519 could be explained by higher relative contributions of epidote and carbonate in bedloads  
520 and muscovite/K-feldspar in suspended loads.

521

#### 522 **5.4. Which minerals control Pb isotopes of river sediments?**

523 To perform the Monte-Carlo simulation for Pb isotopes, we used the data already  
524 published by Garçon et al. (2013b) but we list them for convenience in Table 1. Because  
525 we did not measure the Pb isotopic composition of all the mineral fractions, we used an  
526 estimated value for the “heavy mineral” fraction, containing zircon, monazite and allanite,  
527 to perform our simulation (i.e.  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio of about 800-900 after Garçon et al.,  
528 2013b). As we obtained similar results for  $^{208}\text{Pb}/^{204}\text{Pb}$ ,  $^{207}\text{Pb}/^{204}\text{Pb}$  and  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios,  
529 only the results obtained for  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios are shown in Figure 9. Seven minerals

530 (biotite, magnetite, titanite, amphibole, epidote, garnet and carbonate) always contribute  
531 to less than 10% to the Pb isotopic budget and are not shown in Figure 9. In contrast, clay,  
532 K-feldspar, muscovite, plagioclase and zircon/monazite/allanite contribute significantly  
533 to the final isotopic composition. The modeled  $^{206}\text{Pb}/^{204}\text{Pb}$  cover a very large range of  
534 values between 19 and 55, the highest values being associated with high contributions of  
535 zircon, monazite and allanite in the mixture (Figure 9). Indeed, if we perform a simulation  
536 without these heavy minerals, the modeled  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios only range between 19.0  
537 and 19.8. This is because zircon, monazite and allanite are extremely radiogenic  
538 (estimated  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio of 800-900; Garçon et al., 2013b), and even a minute amount  
539 of this mineral mixture in the sediment ( $\leq 1\text{wt}\%$ ) significantly shifts the modeled Pb  
540 isotopic compositions towards very radiogenic values (Figure 9).

541         The ranges of  $^{206}\text{Pb}/^{204}\text{Pb}$  measured for bedloads (20.6-22.2) and suspended loads  
542 (19.8-20.1) are shown using grey bands in Figure 9. Our results show that for both types  
543 of sediments, the Pb-rich clay and K-feldspar control up to 80% of the Pb isotopic budget  
544 while muscovite and plagioclase are other significant contributors. Our simulations also  
545 show that zircon, monazite and allanite play a key role in increasing the Pb isotopic  
546 compositions of both bedloads and suspended loads. Indeed, if these heavy minerals were  
547 not present, the  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio of any mixture could not exceed 19.8, a value  
548 significantly lower than what is measured on both bedloads and suspended loads. This  
549 implies that the Pb isotopic budget of both bedloads and suspended loads are influenced  
550 by a “heavy mineral effect”, with a stronger impact on bedloads than on suspended loads  
551 (see Figure 9 and Garçon et al. (2013b) for further discussion).

552



## 553 **5.5. Geological implications**

554 We showed above that the difference in Hf, Sr and Pb isotopic compositions  
555 between bedloads and suspended loads sampled at the same location are due to different  
556 mineral contributions that are ultimately controlled by mineral sorting processes within  
557 the water column. We now evaluate the implications of these results for (1) sediment  
558 provenance studies based on isotope data and (2) large-scale isotopic partitioning  
559 between continental and oceanic sediments.

560

### 561 **5.5.1. Sediment provenance studies**

562 Isotopic data are often used to trace erosion distribution in watersheds (e.g., Galy  
563 and France-Lanord, 2001; Clift et al., 2002; Singh and France-Lanord, 2002; Singh et al.,  
564 2008) as well as changes through time of the source of sedimentary material in relation  
565 to changes of tectonic regimes and/or climate (e.g. Walter et al., 2000; Gingele et al., 2007;  
566 Yan et al., 2007; Haley et al., 2008; Singh et al., 2008; Cina et al., 2009; Rahaman et al.,  
567 2009; Stumpf et al., 2011; Hu et al., 2012). They are also largely used in studies focusing  
568 on the changes of ocean circulation through time (Revel et al., 1996; Innocent et al., 1997;  
569 Stille et al., 1997; Bayon et al., 2002; Frank, 2002; Goldstein and Hemming, 2003;  
570 Piotrowski et al., 2004; Gourlan et al., 2008; Haley et al., 2008; Ehlert et al., 2011) or in  
571 studies that attempt to establish average isotopic compositions or ages for large areas of  
572 continental crust (Goldstein et al., 1984; Goldstein and Jacobsen, 1988; Asmerom and  
573 Jacobsen, 1993; Allègre et al., 1996; Millot et al., 2004; Kamber et al., 2005; Dhuime et al.,  
574 2011). It is therefore important to know which isotopic systems are robust tracers of  
575 source compositions and in which conditions.

576 Nd isotopes are certainly an important source tracer. Our study of bedloads,  
577 suspended loads and individual minerals confirms that it is robust. More specifically, we  
578 demonstrate that the isotopic budget of both suspended loads and bedloads is buffered  
579 by monazite and allanite even though these minerals are present in extremely small  
580 proportions (< 0.5 wt.%) in the sediments. Both types of sediments can be used for  
581 provenance studies. We also confirm the conclusions presented by Garçon et al. (2011)  
582 who demonstrated that, in beach placers, monazite by itself was a good proxy to estimate  
583 the Nd isotopic composition of large continental areas. Analyses of populations of  
584 monazite grains or analyses of single grains could be an alternative to whole rock  
585 measurements.

586 Hf isotopic analysis of zircon grains is another widely used approach to trace source  
587 compositions (Bodet and Schärer, 2000; Davis et al., 2005; Harrison et al., 2005; Veevers  
588 et al., 2005; Flowerdew et al., 2006; Hawkesworth and Kemp, 2006; Kemp et al., 2007;  
589 Zeh et al., 2007; Belousova et al., 2010). Using the  $^{176}\text{Hf}/^{177}\text{Hf}$  of bulk sediments as source  
590 tracer is definitively trickier in view of the “zircon effect” highlighted by previous studies  
591 (Patchett et al., 1984; Vervoort et al., 1999; Chauvel et al., 2008; Bayon et al., 2009;  
592 Carpentier et al., 2009; Vervoort et al., 2011). However, our study demonstrates and  
593 quantifies the fact that since zircon almost entirely controls the  $^{176}\text{Hf}/^{177}\text{Hf}$  ratio of  
594 bedloads, these sediments can be used for source provenance studies. In contrast,  
595 suspended loads that are depleted in zircon and have their Hf isotopic budget controlled  
596 by radiogenic minerals such as biotite, muscovite, clay, or amphibole, have isotopic  
597 compositions that differ from that of their source and cannot be used to trace sediment  
598 provenance.

599 A surprising feature is that heavy minerals not only affect the Hf isotopic  
600 compositions of river sediments but also have dramatic effects on their Pb isotopic

601 compositions. Indeed, our calculations indicate that, because of their extremely  
602 radiogenic Pb isotopes, zircon, monazite and allanite have the potential to significantly  
603 increase the Pb isotopic ratios of both bedloads and suspended loads (e.g., as high as 60  
604 for  $^{206}\text{Pb}/^{204}\text{Pb}$ ; Figure 9). Garçon et al. (2013) have shown that mineralogical sorting  
605 processes during fluvial transport lead to an excess concentration of heavy minerals in  
606 bedloads and, to lesser extent, in suspended loads. This “heavy mineral effect” shifts the  
607 Pb isotopic compositions of most river sediments, including some suspended loads,  
608 towards more radiogenic values than their source areas. The best approach to constrain  
609 the Pb isotopic composition of the source areas is thus to concentrate on K-feldspars or  
610 clays separated from sediments because these fractions contain most of the lead initially  
611 present in crustal rocks and are almost not affected by the “heavy mineral effect” (see  
612 Garçon et al., 2013b for further discussion).

613 Finally, using Sr isotopes in river sediments to trace the composition of source areas  
614 appears very difficult without taking into account potential bias due to mineralogical  
615 sorting processes during sediment transport. Bedloads and suspended loads have  
616 different isotopic compositions and, according to our calculations, the more radiogenic Sr  
617 signature of suspended loads compared to bedloads probably comes from higher  
618 contributions of radiogenic muscovite and/or K-feldspar in suspended loads (Figure 7)  
619 together with higher contributions of less radiogenic epidote and carbonate in bedloads.  
620 It remains unclear whether either bedloads or suspended loads have compositions  
621 similar to that of the source areas. As a consequence, Sr isotopes can only be used as a  
622 source proxy if the sources are isotopically extremely different (i.e. range larger than that  
623 created by mineralogical sorting processes), as is rarely the case but occurs in the  
624 Himalayan system.

625

626           **5.5.2. Difference between what goes into the deep ocean and what**  
627 **remains on continents**

628           At a global scale, mineral sorting processes are responsible for significant isotopic  
629 fractionation between continental sediments and terrigenous oceanic clays. Patchett et  
630 al. (1984) were the first to discuss such effects and showed their impact on the Hf isotopic  
631 systematics of sediments. They argued that the “zircon effect”, i.e. retention of  
632 unradiogenic Hf-rich zircons on continents, led to the delivery into the oceans of finer  
633 detrital material, poor in zircons and with radiogenic Hf isotopes. Numerous other studies  
634 confirmed the observation since 1984 (Chauvel et al., 2008; Bayon et al., 2009; Carpentier  
635 et al., 2009; Chauvel et al., 2009; Vervoort et al., 2011). Recently, Garçon et al. (2012;  
636 2013a) showed that almost half of the bias towards radiogenic Hf isotopic seen today in  
637 terrigenous oceanic clays is observed in suspended river sediments on continents. The  
638 Hf Monte-Carlo simulation presented in Figure 5 confirms that suspended loads delivered  
639 at the mouth of a river as large as the Ganga still contain zircons. In particular, we show  
640 that, even if radiogenic minerals such as phyllosilicates contribute to a non-negligible part  
641 of the Hf isotopic composition of the Ganga suspended loads, zircon can still control up to  
642 50% of their Hf isotopic budget (Figure 5). Thus, further sorting must occur in the oceanic  
643 environment to reach the maturity of terrigenous oceanic clay and their characteristic  
644 highly radiogenic zircon-free Hf signature. This sorting could occur in the ocean, next to  
645 the continental margins due to a rapid change of the hydrodynamic conditions (Lisitzin,  
646 1996; Stummeyer et al., 2002).

647           Deposition of heavy minerals in the coastal zone is probably a key process that not  
648 only affects the Hf isotopic composition of sedimentary material delivered to the ocean,  
649 but also affects their Pb and Sr isotopes in a similar way. Our simulations demonstrate  
650 that the Pb isotopic composition of bedloads and even that of suspended loads are biased

651 towards radiogenic values by a “heavy mineral effect”. Thus, we expect terrigenous  
652 oceanic clays to be characterized by less radiogenic Pb isotopic ratios than river  
653 suspended loads. Similarly, the deposition of dense epidote and/or coarse particles in  
654 coastal zones should lead to the delivery into the deep oceans of materials with more  
655 radiogenic Sr isotopic compositions than river suspended loads. Such a potential Sr  
656 isotopic fractionation at the land/ocean interface has already been pointed out by  
657 Eisenhauer et al. (1999) and our calculations confirm their suggestions. Finally,  
658 deposition of heavy monazite and allanite in the coastal zone should not modify the Nd  
659 isotopic composition of sediments delivered in the deep ocean, since these two minerals  
660 have essentially the same  $^{147}\text{Sm}/^{144}\text{Nd}$  as both bulk sediments and continental sources  
661 (Patchett et al., 1984; McLennan, 1989; Garçon et al., 2011; Vervoort et al., 2011).

## 662 **6. Summary and Conclusions**

663 The Monte-Carlo simulations performed in this study provide a framework to  
664 decipher the contribution of individual mineral species to the Nd, Hf, Sr and Pb isotopic  
665 budget of river sediments. They also provide clues to understand the isotopic differences  
666 observed between bedloads and suspended loads that are related to mineral sorting  
667 processes during the fluvial transport of sediments. The results of the Monte-Carlo  
668 simulations are well constrained and could be used to understand the isotopic  
669 systematics of other sediments from large river systems all over the world.

670 An important result is that, despite the diversity of mineral species present in river  
671 sediments, only a few minerals really participate in the Nd, Hf, Sr and Pb isotopic budgets.  
672 Nd isotopes of both bedloads and suspended loads are mainly controlled by monazite and  
673 allanite and they are not modified by mineral sorting processes. Hence, they can be used  
674 to trace the continental sources of sediments. By contrast, mineral sorting processes lead

675 to significant Hf, Sr and Pb isotopic diversity. We demonstrate that the Hf isotopic budget  
676 of bedloads is totally dominated by the contribution of unradiogenic Hf-rich zircons. The  
677 more radiogenic Hf signature of suspended loads results from lower but still significant  
678 contribution of zircons combined to higher contributions of more radiogenic minerals  
679 such as phyllosilicates. The Pb isotopic budget of all river sediments is dominated by K-  
680 feldspar and clay contributions but it is also significantly affected by a “heavy mineral  
681 effect” that deviates the Pb isotopic compositions towards very radiogenic values. Sr  
682 isotopes in river sediments are more difficult to model but they are mainly controlled by  
683 K-feldspar, plagioclase, muscovite, carbonate, clay and epidote with a systematic  
684 difference between bedloads and suspended loads.

685         Because isotopic fractionations caused by mineral sorting processes during  
686 sediment transport can be very large, we stress the need for caution when using the  
687 isotopic compositions of river sediments as provenance proxies. Of the four isotopic  
688 systems studied here, Nd is this only one that is not affected by the mineralogical effects.  
689 We anticipate that hydrodynamic sorting in the coastal zone should further fractionate  
690 the Hf, Sr and Pb isotopic systems in terrigenous oceanic sediments. In particular, the  
691 deposition of heavy minerals at the river/ocean interface could lead to the delivery into  
692 the deep ocean of sediments with even higher  $^{176}\text{Hf}/^{177}\text{Hf}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  and lower Pb  
693 isotopic ratios than river suspended loads. Further studies on the isotopic variability of  
694 terrigenous oceanic sediments are needed to better constrain the impact of mineral  
695 sorting in the oceanic environment and its role on the decoupling of isotopic systems at a  
696 large scale.

697

## 698 **Acknowledgments**

699           We would like to thank A. Galy, V. Galy and M. Lupker for sample collection, S. Andò  
700 for his help during mineral separation, S. Bureau and C. Poggi for their help in the clean  
701 lab, P. Telouk and P. Nonnotte for assistance during isotopic measurements in Lyon and  
702 Brest, as well as N.T. Arndt and E. Lewin for constructive discussions that helped to  
703 develop and interpret the results of the Monte-Carlo simulations. We also greatly thank  
704 the editor Laurie Reisberg and the two reviewers, Jeff Vervoort and Bill White, who made  
705 very constructive comments that certainly improve the content of this manuscript. This  
706 study was supported by fundings from CNRS and INSU programs.

707

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- 958

## 959 **Tables**

960 **Table 1:** Trace element concentrations (ppm) and Nd-Hf-Sr-Pb isotopes of the mineral  
961 fractions separated from the Ganga sediments.

962 *Footnotes:* \* Data already published by Garçon et al. (2013b).  $\pm 2\sigma$  are in-run errors.

963

**Table 1: Trace element concentrations (ppm) and Nd-Hf-Sr-Pb isotopic compositions of the mineral fractions separated from the Ganga sediments.**

Sample Name	BR 717 Msc	BR 717 Bio	BR 717 Fd-K	BR 717 Fd-K Dup	BR 717 Pig	BR 717 Qtz	BR 717 Mag	BR 717 Ep	BR 717 Grt	BR 717 Tn	BR 717 Amp	BR 717 Carb	BR 717 Zrn	BR 717 Mnz	BR 717 Ap	BGP 6 A
Type of mineral	Muscovite	Biotite	K-Feldspar	K-Feldspar (Duplicate)	Plagioclase	Quartz	Magnetite	Epidote	Garnet	Titanite	Amphibole	Carbonate	Zircon	Rich in Monazite and Allanite	Rich in Apatite	Clay
Cs	15.4	65.9	8.94		0.838	0.209	0.579	0.267	0.153	0.557	1.80	0.00913	0.214	0.935	0.504	17.0
Rb	311	435	313		31.1	3.34	7.37	2.54	1.50	9.73	23.3	0.846	3.03	12.0	9.16	167
Ba	967	478	1393		146	19.8	33.0	14.9	3.49	39.9	60.2	16.6	7.01	71.3	427	373
Th	4.58	24.9	4.10		2.37	2.91	13.7	17.8	15.6	151	24.0	1.17	219	2374	28.8	62.0
U	1.54	2.36	0.663		0.622	0.83	3.61	6.49	3.49	50.1	7.40	0.522	612	400	28.9	8.55
Nb	36.1	87.3	0.992		1.39	0.581	20.8	3.69	3.25	229	403	< DL	171	1068	119	16.7
Ta	2.78	9.58	0.112		0.106	0.0304	1.66	0.267	0.42	31.0	43.5	0.013	17.9	162	8.24	1.49
La	9.99	60.9	4.41		3.74	5.23	19.8	61.2	40.4	172	32.6	6.89	32.8	3763	71.1	1021
Ce	27.5	110	14.3		14.4	11.3	41.1	134	91.0	527	71.6	14.7	113	7785	206	206
Pr	2.34	18.2	1.07		0.933	1.29	4.58	17.2	9.79	93.1	8.76	1.95	16.3	883	30.3	24.3
Pb	11.1	4.96	75.1		12.6	3.89	19.1	46.9	0.916	5.52	24.1	7.28	19.3	81.9	7.98	95.2
Nd	8.29	32.0	3.77		3.57	4.59	16.6	71.8	34.5	414	34.1	8.25	79.6	3180	146	90.5
Sr	32.3	7.01	243		195	70.6	11.8	1509	3.31	52.9	38.4	5.26	16.9	274	197	47.9
Sm	1.57	4.77	0.837		0.684	1.04	3.17	17.6	7.83	105	7.88	2.07	32.7	670	54.0	17.3
Zr	210	264	21.9		94.4	27.1	265	183	251	2128	539	< DL	416415	20849	4240	447
Hf	5.70	7.30	0.658		2.56	0.749	7.01	4.76	6.95	72.8	15.5	< DL	10171	639	119	12.2
Ti	4898	16573	185		273	71.4	17180	2706	770	32279	168099	2.29	41321	107327	18799	6416
Eu	0.253	0.416	0.744		0.30	0.176	0.54	5.75	1.01	19.4	1.73	0.523	7.19	59.8	6.20	2.70
Gd	1.25	3.41	0.733		0.585	0.921	2.74	18.2	16.4	100	8.96	2.37	65.1	494	74.3	14.3
Tb	0.185	0.462	0.118		0.0907	0.155	0.46	2.94	5.95	17.4	1.78	0.414	17.8	61.5	14.1	2.02
Dy	1.14	2.59	0.658		0.537	1.01	2.86	18.6	60.9	113	13.3	2.59	178	310	88.5	11.4
Ho	0.226	0.474	0.127		0.108	0.214	0.612	3.78	15.7	22.0	2.83	0.577	55.7	52.5	16.2	2.12
Y	6.49	13.6	3.77		3.39	6.65	17.0	110	379	536	86.1	19.4	1463	1148	432	58.0
Er	0.677	1.32	0.345		0.335	0.645	1.83	10.6	52.0	62.7	8.42	1.76	243	140	42.3	5.84
Lu	24.6	394	11.8		0.461	5.96	3.69	3.42	14.3	8.78	26.2	0.889	15.7	19.8	13.3	40.8
Yb	0.685	1.22	0.322		0.381	0.587	1.92	9.61	58.0	58.9	8.50	1.82	424	140	33.3	5.21
Re-run	0.198	0.192	0.0478		0.062	0.0861	0.296	1.39	8.54	7.91	1.22	0.283	83.5	21.3	4.47	0.738
Sc	13.3	27.3	0.655		0.917	8.14	90.1	102	33.8	45.8	32.5	1.95	325	251	32.0	19.7
V	171	243	1.89		6.86	8.08	1281	414	54.4	139	943	< DL	99.2	408	128	220
Cr	91.3	144	2.74		5.13	3.34	1950	131	76.1	44.8	197	2.00	70.7	236	81.1	143
Co	2.76	47.9	0.691		0.907	0.269	51.0	2.69	23.8	0.683	44.6	0.956	0.443	2.83	1.25	27.3
Ni	6.39	79.7	2.77		3.62	0.464	170	6.34	1.55	3.28	35.6	1.40	3.86	4.74	2.50	99.0
<sup>143</sup> Nd/ <sup>144</sup> Nd ± 2σ	0.511844 ± 15	0.511834 ± 11	0.511834 ± 19		0.511901 ± 33		0.512089 ± 9	0.512235 ± 10	0.511906 ± 11	0.512182 ± 9	0.511929 ± 24	0.511843 ± 10	0.512062 ± 10	0.511732 ± 8	0.511921 ± 7	0.511827 ± 10
Re-run					0.511820 ± 9									0.511741 ± 6		
<sup>176</sup> Hf/ <sup>177</sup> Hf ± 2σ	0.282218 ± 9	0.282162 ± 11	0.282400 ± 15	0.282313 ± 15	0.282325 ± 9		0.282602 ± 8	0.282803 ± 12	0.282698 ± 5	0.282955 ± 16	0.282236 ± 8		0.281970 ± 5		0.282234 ± 8	
<sup>87</sup> Sr/ <sup>86</sup> Sr ± 2σ	0.831306 ± 10	0.812332 ± 8	0.758656 ± 8	0.755512 ± 7	0.722744 ± 8		0.728827 ± 7	0.712509 ± 7	0.746224 ± 8	0.722704 ± 7	0.725748 ± 8	0.713942 ± 8	0.734440 ± 20		0.746642 ± 8	
<sup>208</sup> Pb/ <sup>206</sup> Pb ± 2σ *	20.2008 ± 16	20.5508 ± 26	19.3257 ± 16	19.3318 ± 12	18.9724 ± 18		19.5343 ± 14	18.9028 ± 14		31.4562 ± 22	19.9482 ± 12	19.9809 ± 8			18.8658 ± 12	

964 **Table 2:** Sr isotopic compositions of bedloads and suspended loads sampled at the  
965 outflow of the Ganga River.

966 *Footnotes:* \* These Sr isotopic compositions were analyzed on the silicate fractions of the  
967 sediments (i.e. after a leaching in 1N acetic acid) in Nancy, France, following the methods  
968 described by Galy et al. (1999). DL stands for detection limit.  $\pm 2\sigma$  are in-run errors.

969

970

**Table 2:** Sr isotopes in bedloads and suspended loads sampled at the outflow of the Ganga River

Sample Name	Latitude	Longitude	Sampling date	Type of sediment	Sampling depth (m)	$^{86}\text{Sr}/^{87}\text{Sr}$	$\pm 2\sigma$	Percentage of the bulk sample (%)	Sr (ppm)
BGP 6	24.35833	88.60833	2-Aug-93	Bedload		0.758885	8		115
BR 414	24.05290	89.02465	13-Jul-04	Suspended Load	2	0.773743	8		91.0
BR 412	24.05290	89.02465	13-Jul-04	Suspended Load	6.5	0.768185	12		101
BR 418	24.05290	89.02465	13-Jul-04	Bedload	10	0.754593	8		103
BR 717	24.05290	89.02465	17-Aug-07	Bedload	11	0.757160	8		98.3
BR 522*	24.04187	89.0362	23-Jul-05	Suspended load	0	0.771110	10		95.8
BR 522 50-200 $\mu\text{m}^*$								4.6	98.3
BR 522 20-50 $\mu\text{m}^*$						0.755340	10	42.8	99.3
BR 522 2-20 $\mu\text{m}^*$						0.782400	10	42.2	83.0
BR 522 0.2-2 $\mu\text{m}^*$						0.757570	10	10.3	48.4



971 **Figures**

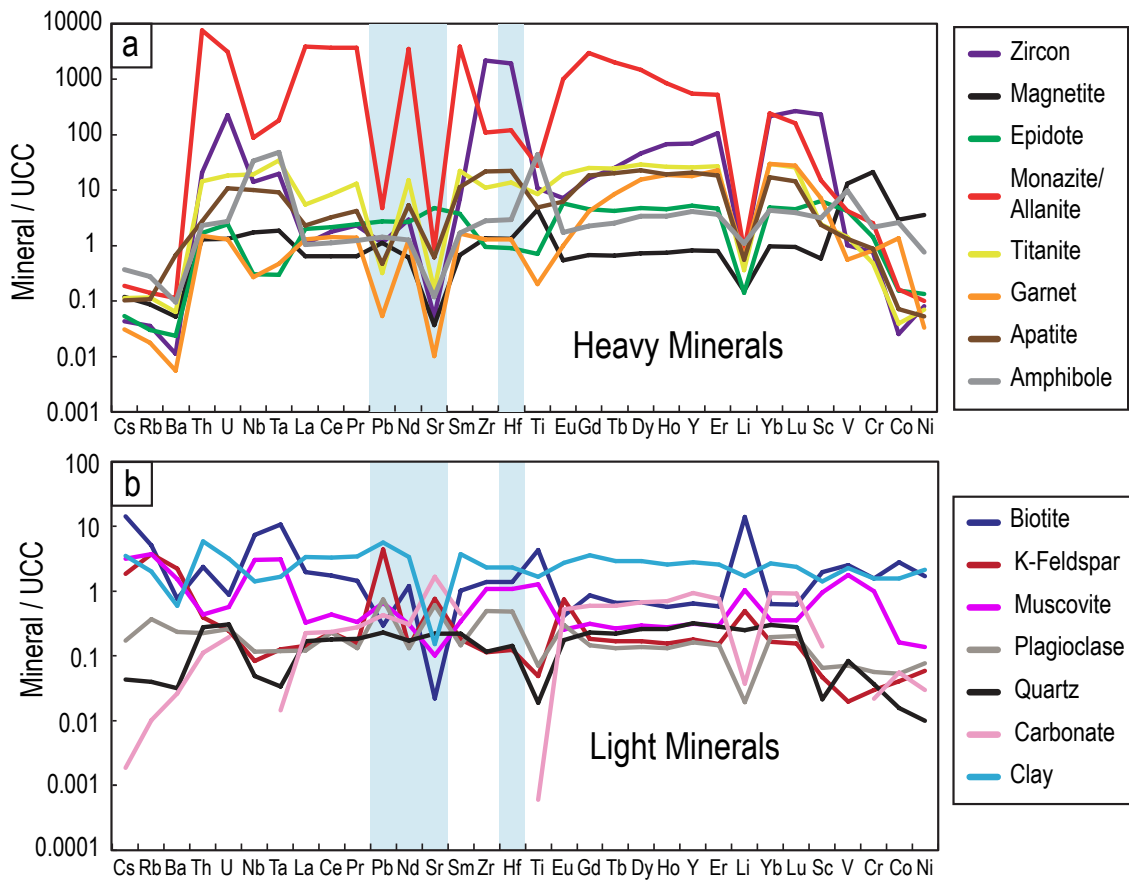
972 **Figure 1:** Map of the Bangladesh delta showing sampling sites for river sediments.

973 **Figure 1**



986 **Figure 2:** Trace element patterns normalized to the average composition of the upper  
 987 continental crust (UCC) of Rudnick and Gao (2003). **a)** Heavy minerals. **b)** Light minerals  
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 989

**Figure 2**



990 **Figure 3:** Results of the Monte-Carlo simulation for Nd isotopes.

991 Proportions of minerals used to perform the Monte-Carlo simulation are shown in the

992 histogram in the top right corner of the Figure (see Supplementary File for more details).

993 Color dots indicate the contributions of the different minerals to the Nd isotopic

994 compositions of the 100,000 modeled mixtures. The contributions of carbonate, garnet,

995 amphibole, epidote, apatite, zircon, magnetite, muscovite, plagioclase, and K-feldspar are

996 not shown in this figure because they represent less than 10% of the whole range of

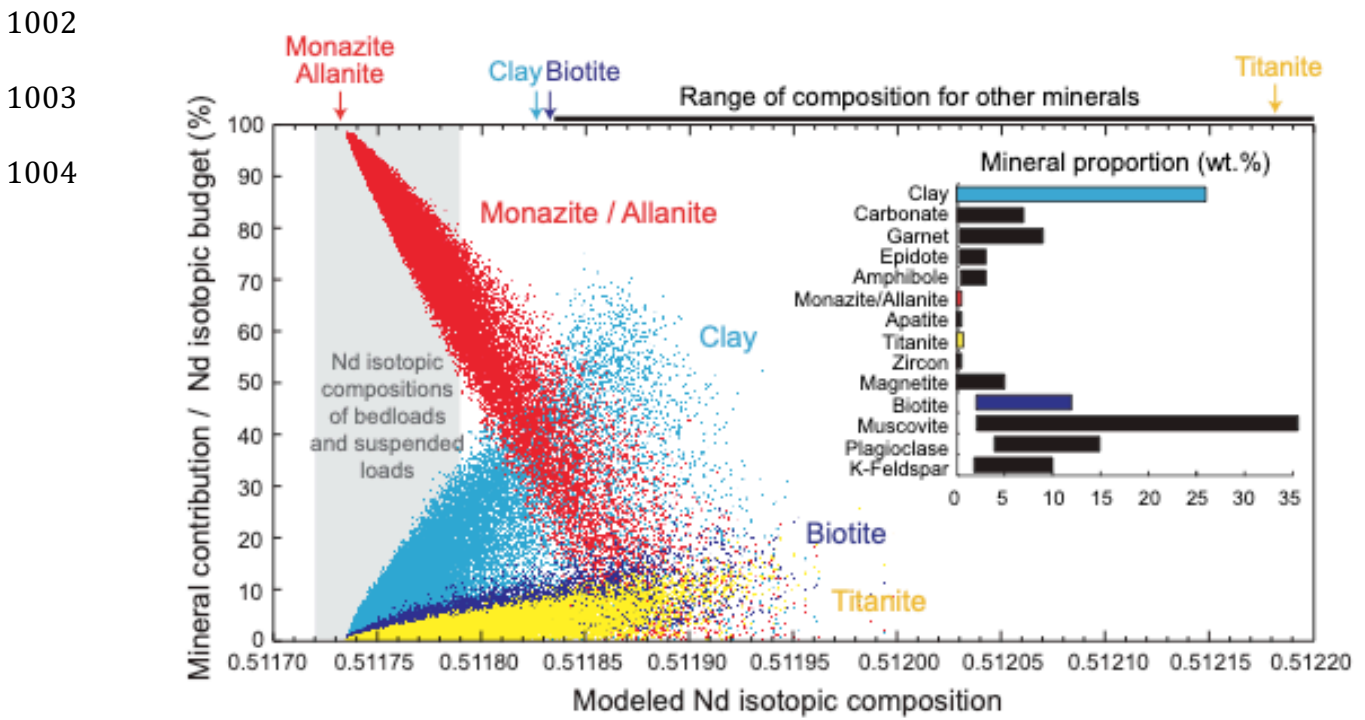
997 modeled Nd isotopic compositions. The vertical light grey band shows the range of Nd

998 isotopic compositions measured in bedloads and suspended loads sampled at the outflow

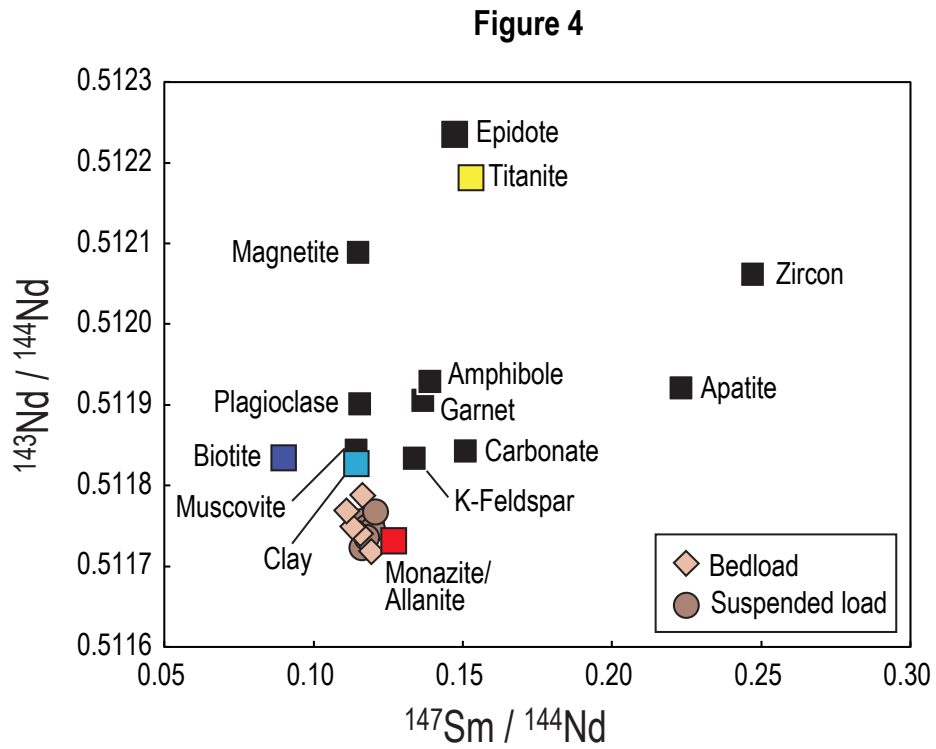
999 of the Ganga River and was constructed using the data of Garçon et al. (2013a). Arrows

1000 on the top axis indicate isotopic compositions of the most important minerals while the

1001 black line shows the range of isotopic compositions for all other minerals.



1005 **Figure 4:** Variations of  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios against  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios for the different  
 1006 mineral fractions.  
 1007 Bedloads and suspended loads shown in this Figure were sampled at the outflow of the  
 1008 Ganga River. Data are from Garçon et al. (2013a).



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1020 **Figure 5:** Results of the Monte-carlo simulation for Hf isotopes.

1021 Proportions of minerals used to perform the Monte-Carlo simulation are shown in the

1022 histogram in the top right corner of the Figure (see Supplementary File for more details).

1023 Color dots indicate the contributions of the different minerals to the Hf isotopic

1024 compositions of the 100,000 modeled mixtures. The contributions of garnet, epidote,

1025 magnetite, amphibole, plagioclase, and K-feldspar are not shown in this figure because

1026 they represent less than 10% of the whole range of modeled Hf isotopic compositions.

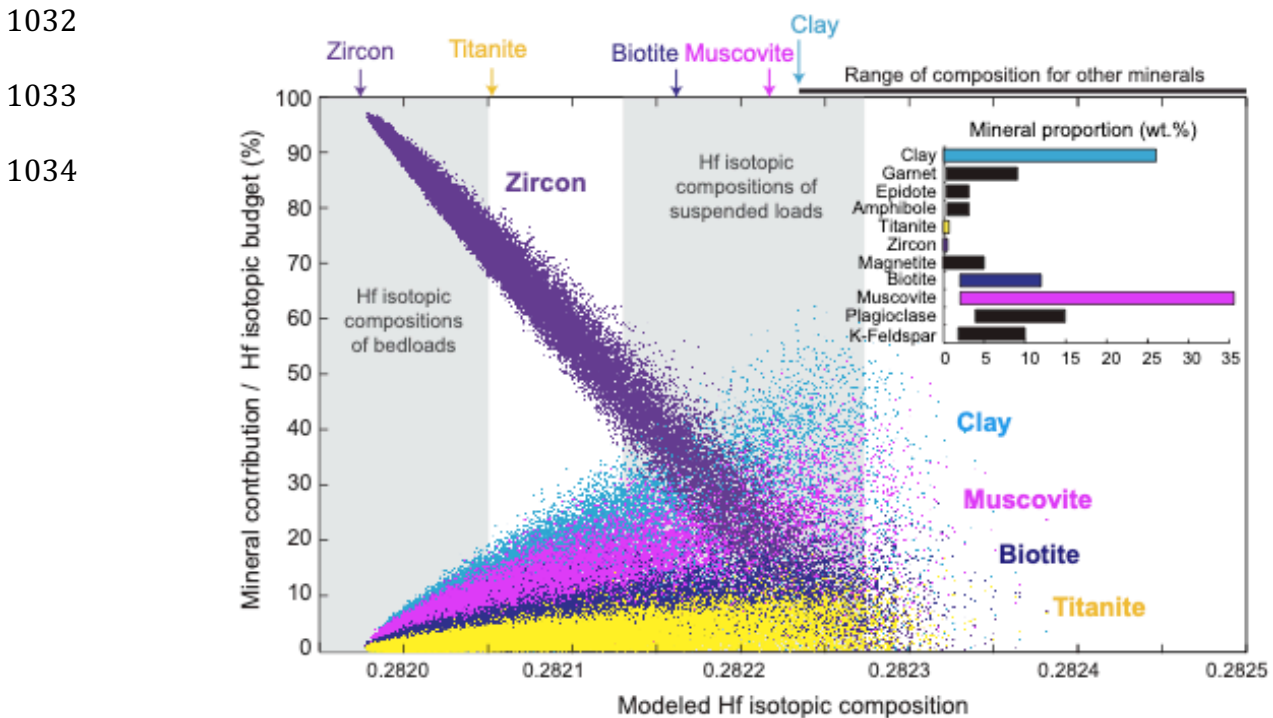
1027 Vertical light grey bands show the range of Hf isotopic compositions measured in

1028 bedloads and suspended loads sampled at the outflow of the Ganga River and were

1029 constructed using the data of Garçon et al. (2013a). Arrows on the top axis indicate

1030 isotopic compositions of the most important minerals while the black line shows the

1031 range of isotopic compositions for all other minerals.



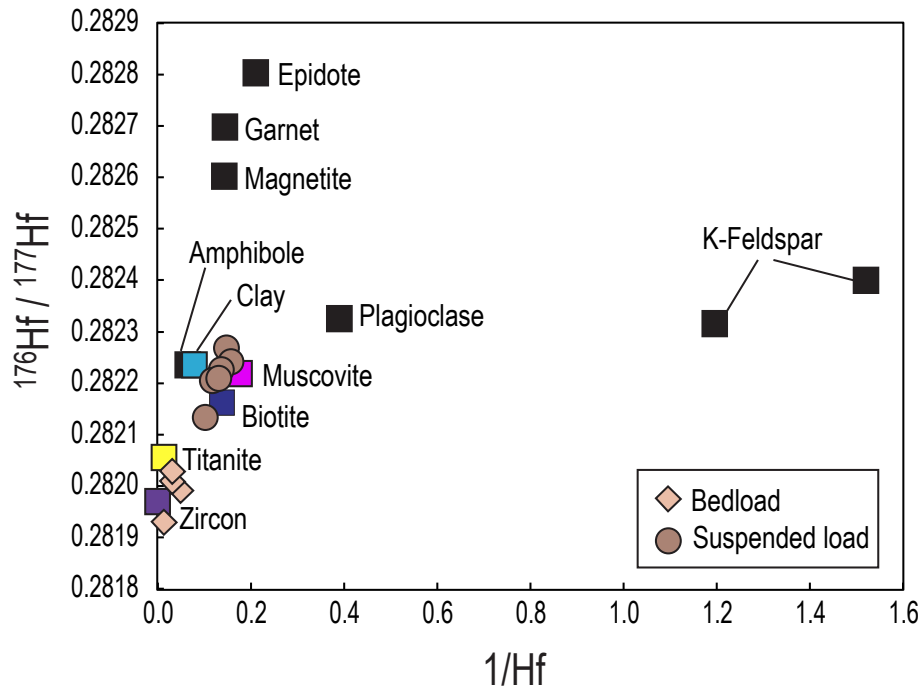
1035 **Figure 6:** Variations of  $^{176}\text{Hf}/^{177}\text{Hf}$  ratios against  $1/\text{Hf}$  ratios for the different mineral  
1036 fractions.

1037 Data for bedloads and suspended loads sampled at the outflow of the Ganga River are  
1038 from Garçon et al. (2013a).  $1/\text{Hf}$  ratios of river sediments have been recalculated on a  
1039 quartz-free basis by normalizing their  $\text{SiO}_2$  contents, as published by Lupker et al. (2011),  
1040 to that of our clay fraction (i.e. 43 wt%). Normalized  $1/\text{Hf}$  ratios were thus calculated  
1041 following this equation:  $(1/\text{Hf})_N = (1/\text{Hf})_{\text{sample}} * (\text{SiO}_2, \text{clay} / \text{SiO}_2, \text{sample})$

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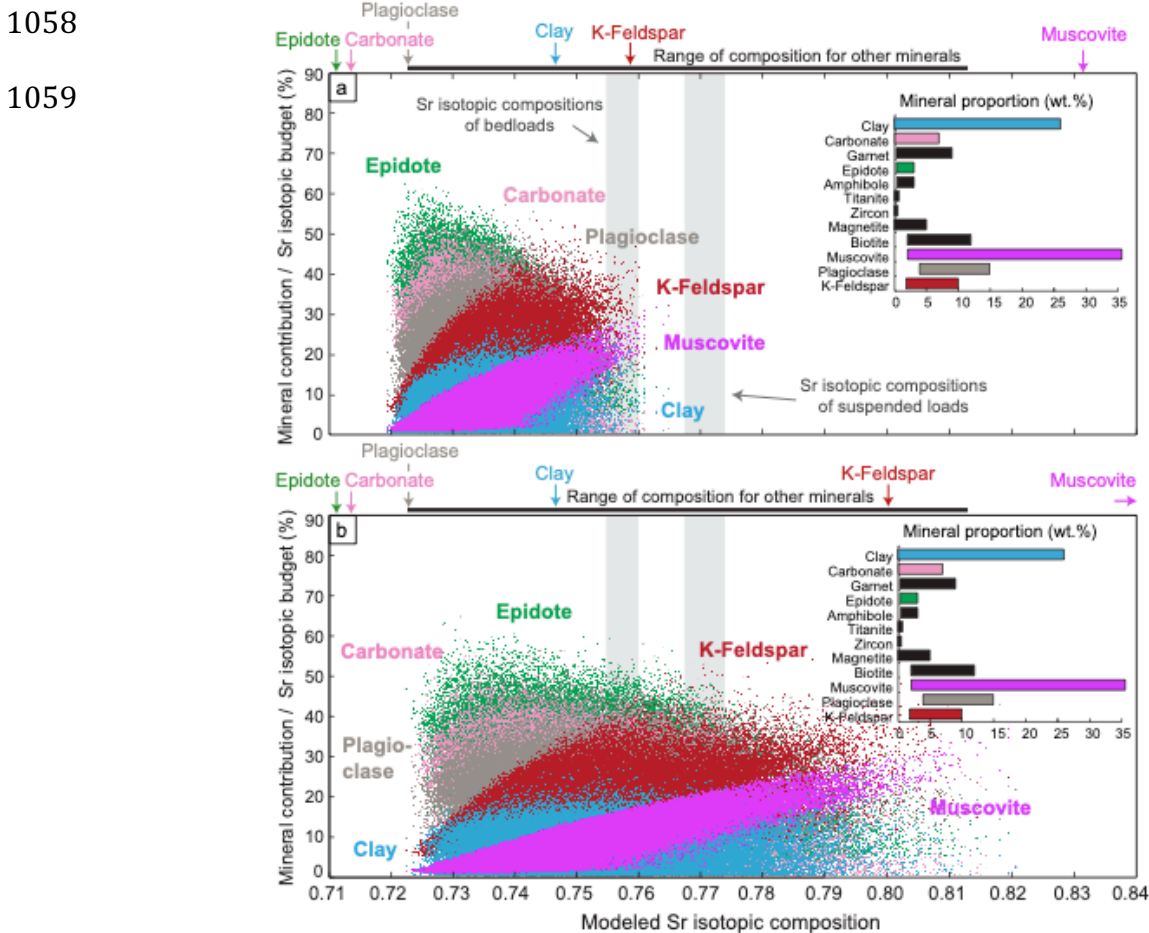
1043

**Figure 6**



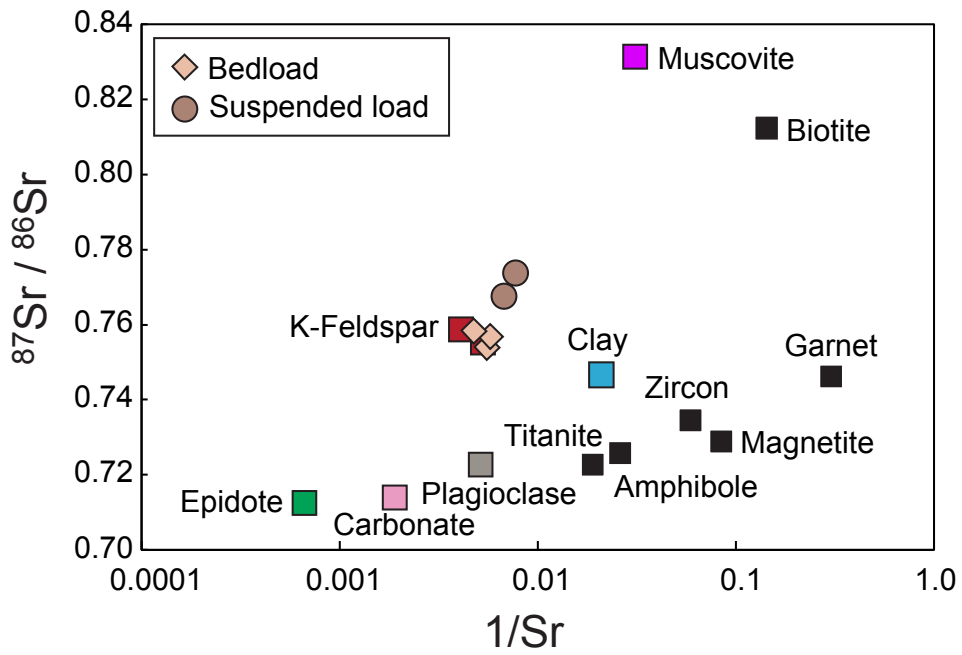
1044 **Figure 7:** Results of the Monte-Carlo simulation for Sr isotopes.

1045 **a)** Monte-Carlo simulation performed using the Sr isotopic compositions as measured in  
1046 our mineral fractions. **b)** Monte-Carlo simulation performed using an  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of  
1047 0.800 for the K-feldspar fraction and 1.000 for the muscovite fraction. Proportions of  
1048 minerals used to perform the Monte-Carlo simulation are shown in the histograms in the  
1049 top right corner of the Figures (see Supplementary File for more details). Color dots  
1050 indicate the contributions of the different minerals to the Sr isotopic compositions of the  
1051 100,000 modeled mixtures. The contributions of garnet, amphibole, titanite, zircon,  
1052 magnetite, and biotite are not shown in this figure because they represent less than 10%  
1053 of the whole range of modeled Sr isotopic compositions. Vertical light grey bands show  
1054 the range of Sr isotopic compositions measured in bedloads and suspended loads sampled  
1055 at the outflow of the Ganga River and analyzed in this study. For each simulation, arrows  
1056 on the top axis indicate isotopic compositions of the most important minerals while the  
1057 black line shows the range of isotopic compositions for all other minerals.



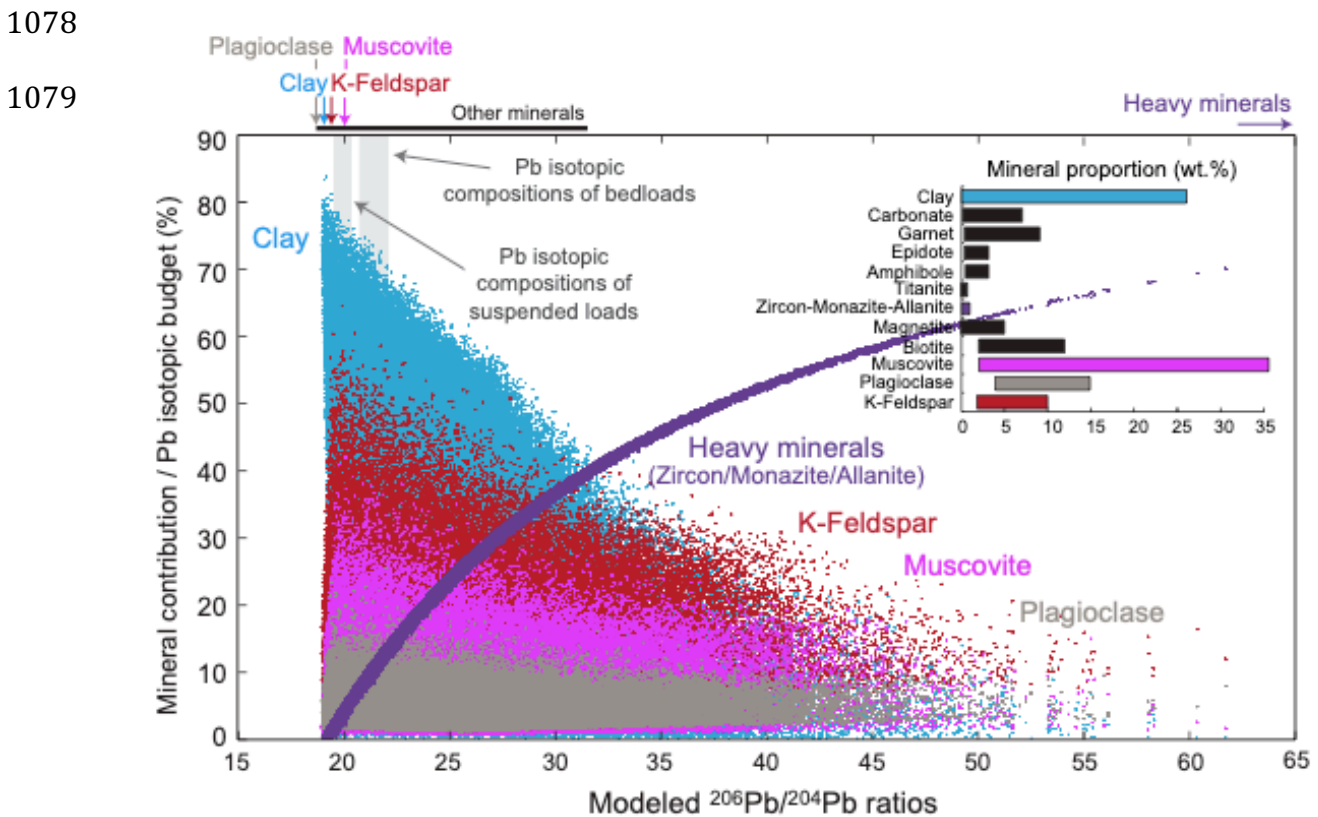
1060 **Figure 8:** Variations of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios against  $1/\text{Sr}$  ratios (logarithmic scale) for the  
 1061 different mineral fractions and the river sediments analyzed in this study. As in Figure 6,  
 1062 we normalized the  $1/\text{Sr}$  ratios of bedloads and suspended loads using their  $\text{SiO}_2$  contents  
 1063 and that of the clay fraction to correct from quartz dilution effects.

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1066 **Figure 9:** Results of the Monte-Carlo simulation for Pb isotopes.  
 1067 Proportions of minerals used to perform the Monte-Carlo simulation are shown in the  
 1068 histograms in the top right corner of the Figures (see Supplementary File for more  
 1069 details). Color dots indicate the contributions of the different minerals to the Pb isotopic  
 1070 compositions of the 100,000 modeled mixtures. The contributions of carbonate, garnet,  
 1071 epidote, amphibole, titanite, magnetite and biotite are not shown in this figure because  
 1072 they represent less than 10% of the whole range of modeled  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios. Vertical  
 1073 light grey bands show the range of Pb isotopic compositions measured in bedloads and  
 1074 suspended loads sampled at the outflow of the Ganga River and were constructed using  
 1075 the data of Garçon et al. (2013b). Arrows on the top axis indicate isotopic compositions of  
 1076 the most important minerals while the black line shows the range of isotopic  
 1077 compositions for all other minerals.



## Supplementary File

### *Input parameters for the Monte-Carlo simulations*

- **Mineral proportions**

Based on the mineral proportions estimated in suspended loads and bedloads by Garzanti et al. (2010, 2011) and Lupker et al. (2012) (see Supplementary Table 1), we defined possible ranges of weight proportions for the most important mineral species in the Ganga river sediments (see Table A below). We combined the albite and Ca-plagioclase estimates from Garzanti et al. (2010; 2011) to determine a range for plagioclase; similarly, we combined the calcite + dolomite estimates from Garzanti et al. (2010; 2011) and Lupker et al. (2012) to estimate the carbonate proportion and we used the opaques + Fe-oxides estimates from Garzanti et al. (2010; 2011) to establish the proportion of magnetite. The first column of Table A shows the range of proportions used for the simulations that are shown in Figures 3, 5, 7 and 9. We deliberately choose to use ranges that are larger than those determined by Garzanti et al (2010, 2011) and Lupker et al. (2012) to insure that our simulations maximize the errors and include extreme scenarios. The second column in Table A provides the ranges used to assess the robustness of the method (see below).

	<b>Range of proportions used for the simulations</b>	<b>Range of proportions used to assess the robustness of the simulations</b>
<b>K-feldspar</b>	2 – 10 wt.%	0 – 20 wt.%
<b>Plagioclase (albite + Ca-plagioclase)</b>	4 – 15 wt.%	0 – 30 wt.%
<b>Muscovite</b>	2 – 36 wt.%	0 – 72 wt.%
<b>Biotite</b>	2 – 12 wt.%	0 – 24 wt.%
<b>Magnetite (opaques + Fe-oxides)</b>	0 – 5 wt.%	0 – 10 wt.%
<b>Zircon</b>	0 – 0.5 wt.%	0 – 1 wt.%
<b>Titanite</b>	0 – 0.7 wt.%	0 – 1.4 wt.%
<b>Apatite</b>	0 – 0.5 wt.%	0 – 1 wt.%
<b>Monazite/Allanite</b>	0 – 0.5 wt.%	0 – 1 wt.%
<b>Amphibole</b>	0.3 – 3.0 wt.%	0 – 6 wt.%
<b>Epidote</b>	0.2 – 3.0 wt.%	0 – 6 wt.%
<b>Garnet</b>	0.2 – 9.0 wt.%	0 – 18 wt.%
<b>Carbonate (calcite + dolomite)</b>	0.5 – 7 wt.%	0 – 14 wt.%
<b>Clay</b>	0 – 26 wt.%	0 – 52 wt.%

**Table A:** Ranges of mineral proportions used for the Monte-Carlo simulations

Some of the minerals identified by Garzanti et al. (2010, 2011) (Supplementary Table 1) are not included in our simulations: quartz, chlorite, tourmaline, rutile, Ti oxide, pyroxene, spinel, chloritoid, staurolite, kyanite, sillimanite, fibrolite, olivine and xenotime. Garzanti et al. (2010, 2011) showed that these minerals contribute to less than 2% in the Nd, Hf, Sr and Pb elemental budgets of the sediments. We therefore chose to minimize the analytical work and did not analyze their trace element contents and isotopic compositions.

- **Concentrations and isotopic compositions of minerals**

We performed the Monte-Carlo simulations using the Nd, Hf, Sr, Pb concentrations and isotopic compositions of mineral separates as listed in Table 1. The only exceptions are the Nd concentrations of monazite and apatite that we took from Garzanti et al. (2010, 2011) (94, 000 ppm and 635 ppm, respectively) because the fractions that we analyzed were not pure enough. In the fraction that we measured, the REE are certainly diluted by the presence of other minerals but we are still confident that the Nd isotopic compositions are representative of the pure minerals. As explained in the main text of the manuscript, only one analysis was performed for each mineral species because of the complexity of the mineral separation procedure and the limited amount of samples available for the study. However, a large number of grains were analyzed in each mineral fraction, meaning that our measurements should be representative of the average compositions of the mineral populations. Errors in the simulations associated to such uncertainties are evaluated below.

### *Monte-Carlo simulations*

For each mineral species, proportions were randomly sampled ( $n=100,000$ ) in uniform distributions within the range of proportions indicated in the Table A (first column). The simulations were designed to allow the sum of the mineral proportions to add up to more or less than 100% because the calculations done in the simulations use relative mineral proportions only, hence this does not influence the final results. We have tested other simulations in which the mineral proportions were rejected and re-sampled if their sums were  $>105\%$  and/or  $<95\%$  but this does not change the results of the simulations. Then, using these proportions together with concentrations and isotopic compositions of individual minerals, we calculated the isotopic compositions of the resulting mixtures.

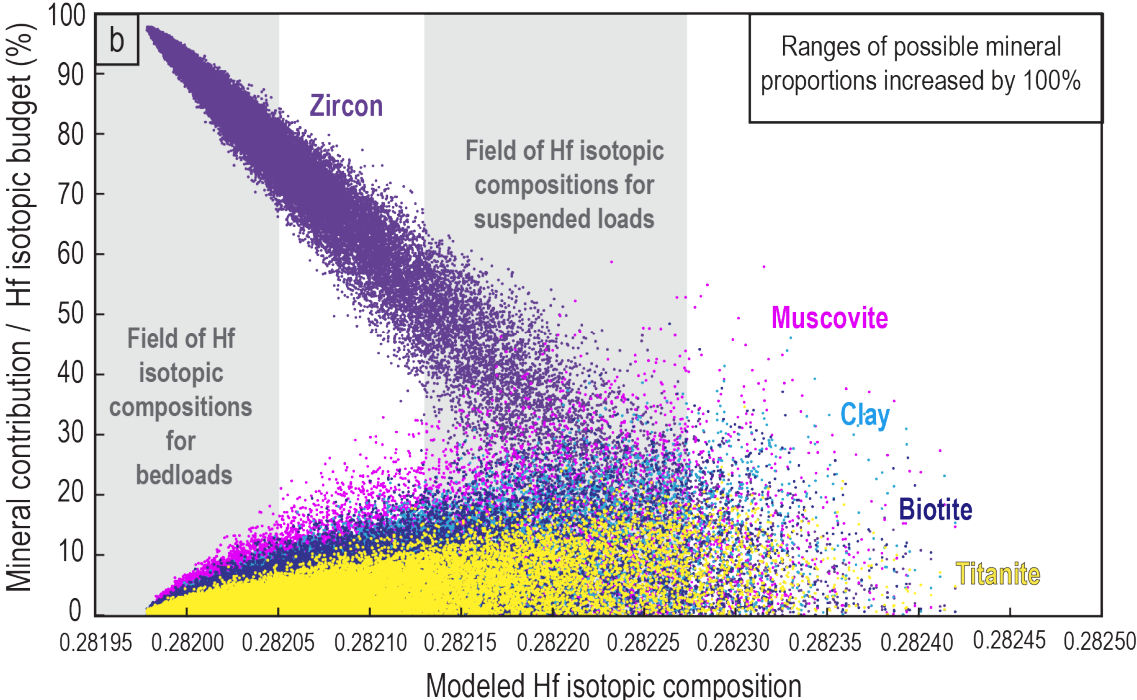
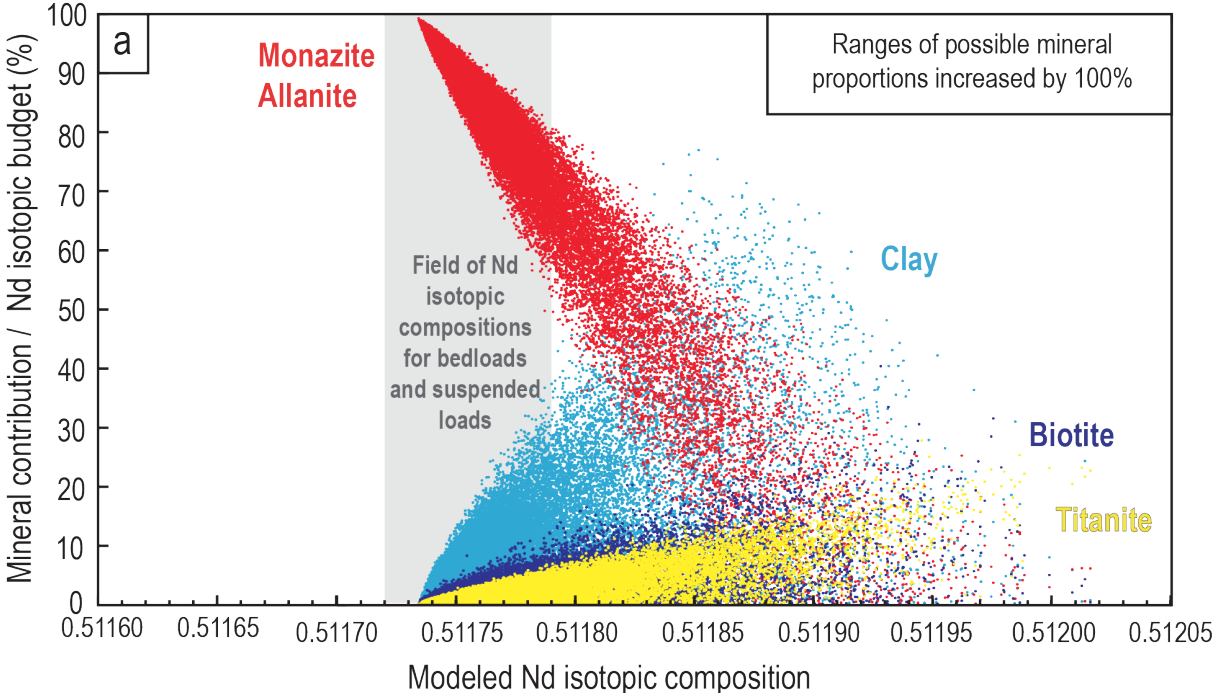
Note that Nd, Hf, Pb and Sr concentrations modeled by this set of Monte-Carlo simulations are not identical to concentrations measured in sediments because (a) the percentages of important minerals are relative and not absolute, and (b) we did not take into account quartz that significantly dilutes all concentrations. This is why we performed other simulations not shown here. They were designed to model only the concentrations in the sediments but not the isotopic compositions. Results showed that the range of Nd, Hf, Sr and Pb concentrations in the Ganga sediments are perfectly reproduced when quartz is added to the minerals listed in Table A.

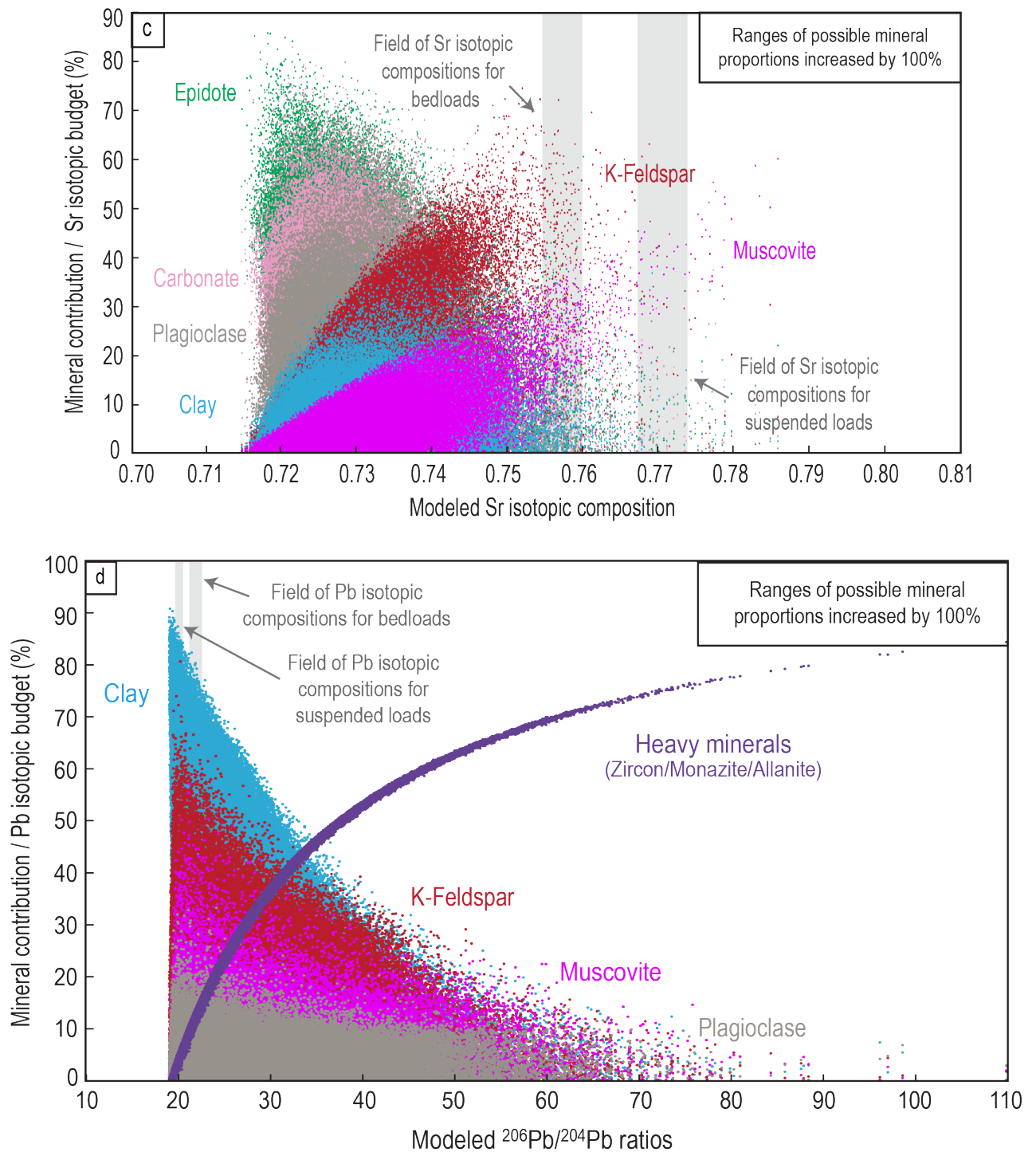
### *Robustness of the Monte-Carlo simulations*

To assess the robustness and uncertainties of the Monte-Carlo simulations we performed several tests. We first increased the ranges of possible mineral proportions by 100% (second column in Table A) to evaluate the sensitivity of the simulations to this parameter. Results are shown below in Figure A for the four isotopic systems. The important point is that the contribution of the different mineral species to the isotopic budgets of the Himalayan sediments remain remarkably similar, showing that the simulation results are well constrained. The mineralogy would need to be completely different for the results to be modified. In turn, this implies that the results presented in this study should be valid for most sediment sampled in large river systems all over the world.

We also tested the sensitivity of the simulations to changes of both concentrations and isotopic compositions of the minerals. To do that, we added an initial step that randomly samples the concentrations and isotopic compositions of each mineral species in given Gaussian distributions centered on the values that we measured in the mineral separates. We deliberately assumed very large errors on the values, with standard deviations ( $2\sigma$ ) of  $\pm 20\%$  for the concentrations,  $\pm 0.0001$  for Nd and

Hf isotopic compositions,  $\pm 0.01$  for Sr isotopic compositions and  $\pm 1\%$  for Pb isotopic compositions. The rest of the calculation is similar to those shown in Figures 3, 5, 7 and 8. The results of these simulations (not shown here) indicate that the contributions of the different mineral species to the isotopic budget of the Ganga sediments still remain the same as in Figures 3, 5, 7 and 8. The uncertainties entailed by our data set are thus limited and we are confident that the results presented in this study would not significantly change even if our data set did not catch all the compositional heterogeneity of the Himalayan minerals.





**Figure A:** Results of the Monte-Carlo simulations when increasing the input ranges of mineral proportions.

a) Nd; b) Hf; c) Sr and d) Pb. These simulations were performed using the same minerals as those considered in Figures 3, 5, 7 and 9 but using proportion ranges much larger (values provided in the second column of Table A). Minerals that contribute to less than 10% of the isotopic budgets are not

shown in the figures. Note that the ranges of modeled Sr isotopic compositions simulations still remain incompatible with the ranges measured for suspended loads from the Ganga River.

**Supplementary Table 1:** Weight mineral proportions in Ganga sediments modified after Garzanti et al. (2010, 2011)

Surface suspended loads (<3m)					Deep suspended loads (>8m)					Bedloads				
	Average	2 $\sigma$	Minimum	Maximum		Average	2 $\sigma$	Minimum	Maximum		Average	2 $\sigma$	Minimum	Maximum
Quartz	34.3	22.8	26.2	42.4	Quartz	48.7	10.6	43.2	54.1	Quartz	55.4	14.9	39.8	66.0
K-feldspar	2.6	1.6	2.1	3.2	K-feldspar	5.7	2.3	4.3	6.9	K-Feldspar	6.6	3.1	3.9	8.5
Albite	4.6	2.4	3.8	5.4	Albite	5.9	4.0	3.9	8.2	Plagioclase	6.0	2.9	2.9	7.4
Ca-plagioclase	2.4	2.3	1.6	3.2	Ca-plagioclase	4.2	2.8	3.5	6.4					
Lithic grain	ND				Lithic grain	ND				Lithic grain	14.5	5.2	10.9	18.6
Calcite	1.2	0.3	1.1	1.3	Calcite	2.9	1.6	1.9	3.7	Calcite*	3.4	2.5	0.5	4.7
Dolomite	1.8	0.2	1.7	1.9	Dolomite	2.3	2.1	1.1	3.5	Dolomite	ND			
Muscovite	17.5	9.2	14.2	20.7	Muscovite	8.9	5.4	5.0	11.0	Micas	9.8	21.9	2.2	36.1
Biotite	10.4	4.3	8.8	11.9	Biotite	5.5	3.0	3.8	7.3					
Chlorite	2.8	1.4	2.4	3.3	Chlorite	1.7	1.1	1.1	2.3	Chlorite	ND			
Clay	19.5	18.6	12.9	26.0	Clay	9.8	3.6	8.2	12.0	Clay	ND			
Fe-oxides	0.2	0.2	0.2	0.3	Fe-oxides	0.1	0.1	0.02	0.1	Fe-oxides	ND			
Fibrolite	ND				Fibrolite	ND				Fibrolite	0.1	0.2	0	0.3
Opagues	1.3	0.1	1.2	1.3	Opagues	1.0	0.7	0.8	1.6	Opagues	1.1	2.8	0.1	3.7
Zircon	0.1	0.02	0.1	0.1	Zircon	0.1	0.3	0.03	0.4	Zircon	0.1	0.4	0	0.5
Tourmaline	0.2	0.1	0.2	0.3	Tourmaline	0.2	0.1	0.2	0.3	Tourmaline	0.3	0.7	0.1	1.1
Rutile	0.2	0.2	0.1	0.3	Rutile	0.2	0.2	0.1	0.3	Rutile	0.1	0.2	0	0.2
Ti oxide	0.4	0.1	0.4	0.4	Ti oxide	0.2	0.2	0.1	0.3	Ti oxide	0.0	0.1	0	0.2
Titanite	0.1	0.1	0.1	0.2	Titanite	0.2	0.3	0.1	0.5	Titanite	0.3	0.5	0.05	0.7
Apatite	0.1	0.04	0.1	0.1	Apatite	0.1	0.1	0.03	0.1	Apatite	0.1	0.4	0	0.5
Monazite	ND				Monazite	ND				Monazite	0.1	0.3	0	0.4
Amphibole	0.5	0.6	0.3	0.7	Amphibole	1.1	0.6	0.9	1.6	Amphibole	1.4	2.0	0.4	3.0
Pyroxene	0.2	0.1	0.2	0.3	Pyroxene	0.3	0.2	0.2	0.4	Pyroxene	0.3	0.5	0.1	0.8
Spinel	ND				Spinel	ND				Cr-spinel	ND		0	0.0
Epidote	1.4	0.8	1.1	1.7	Epidote	1.0	0.3	0.8	1.1	Epidote	1.0	1.5	0.2	2.2
Allanite	ND				Allanite	0.02	0.1	0	0.1	Allanite	ND		0	0.0
Chloritoid	ND				Chloritoid	ND				Chloritoid	ND		0	0.1
Garnet	0.4	0.5	0.3	0.6	Garnet	1.2	1.9	0.5	2.6	Garnet	2.8	6.5	0.2	8.2
Staurolite	ND				Staurolite	0.03	0.02	0.02	0.04	Staurolite	0.1	0.1	0	0.2
Kyanite	0.04	0.1	0.0	0.1	Kyanite	0.1	0.1	0.07	0.19	Kyanite	0.2	0.5	0	0.7
Sillimanite	ND				Sillimanite	0.1	0.1	0.03	0.2	Sillimanite	0.02	0.1	0	0.1
Sum	102.2				Sum	101.4				Sum	103.7			

Minerals taken into account in our Monte-Carlo simulations are highlighted in gray.

ND: not determined or under detection limit

\*not determined by Garzanti et al. (2010; 2011). Values from Lupker et al. (2012)

**Supplementary Table 2:** Trace element concentrations and Nd-Hf-Pb isotopes in sediments sampled at the outflow of the Ganga River from Garçon et al. (2013a; 2013b)

Sample Name	BGP 6	BGP 6 Dup	BR 415	BR 414	BR 413	BR 412	BR 411	BR 418	BR 8252	BR 8253	BR 717
Type of sediment	Bedload	Bedload	Suspended Load	Suspended Load	Suspended Load	Suspended Load	Suspended Load	Bedload	Bedload	Suspended Load	Bedload
Sampling depth (m)			0	2	4	6.5	9	10	14	0	11
Cs	3.83	4.02	15.4	13.6	11.8	11.7	8.08	2.64	3.05	8.92	3.87
Rb	77.9	85.2	202	186	174	175	107	56.6	34.5	51.7	50.3
Ba	294	307	627	562	531	527	372	212	203	240	244
Th	30.2	31.3	20.0	19.0	19.3	18.0	15.1	29.7	24.3	16.1	74.2
U	5.41	5.71	4.64	4.08	4.15	3.96	2.92	5.14	3.33	1.85	11.7
Nb	12.9	12.0	17.4	17.9	16.5	15.7	13.6	18.4	13.6	15.8	20.3
Ta	1.78	1.44	1.54	1.57	1.45	1.39	1.37	2.08	2.91	1.31	2.50
La	67.1	70.2	42.0	41.3	42.2	40.0	32.8	71.0	41.6	30.8	141
Ce	144	145	89.4	88.1	89.2	84.7	76.3	146	110	75.4	292
Pr	15.5	16.4	9.75	9.77	9.87	9.29	7.84	16.7	10.6	8.12	33.1
Pb	17.8	17.6	30.4	28.2	25.3	17.4	15.0	10.7	10.7	10.5	14.2
Nd	56.5	60.8	35.0	35.7	35.8	34.0	29.0	60.4	39.4	31.2	121
Sr	115	117	84.1	91.0	99.3	101	88.6	103	83.6	56.5	98.3
Sm	10.9	11.4	6.93	6.98	7.01	6.77	5.60	12.0	7.63	6.27	22.3
Zr	683	667	178	204	227	199	249	479	403	196	1550
Hf	17.1	17.4	4.73	5.22	5.79	5.17	6.14	12.1	10.5	5.13	40.5
Ti	3155	3166	4137	4198	3976	3833	3314	4355	2523	4323	5299
Eu	1.39	1.45	1.27	1.26	1.23	1.20	1.04	1.48	0.928	1.19	2.24
Gd	8.51	9.10	6.19	6.15	6.05	5.80	4.96	10.1	6.41	5.44	17.7
Tb	1.25	1.29	0.984	0.945	0.93	0.872	0.782	1.64	0.977	0.854	2.52
Dy	7.24	7.61	5.97	5.74	5.72	5.42	4.81	10.3	5.70	5.32	14.7
Ho	1.39	1.48	1.17	1.15	1.13	1.06	0.95	2.12	1.16	1.06	2.83
Y	41.1	43.5	32.7	32.8	32.5	30.9	27.1	62.0	34.5	31.1	85.6
Er	3.99	4.24	3.33	3.30	3.21	3.07	2.65	6.35	3.33	3.04	8.11
Li	14.7	16.3	51.8	48.2	43.5	41.9	32.2	15.8	12.6	45.3	15.7
Yb	3.87	4.16	3.18	3.11	3.06	2.88	2.51	6.23	3.32	2.95	8.17
Lu	0.587	0.614	0.461	0.45	0.448	0.417	0.366	0.905	0.50	0.433	1.25
Sc	7.74	9.81	15.0	14.4	13.5	12.8	9.04	13.2	4.56	11.8	14.3
V	32.7	34.8	93.4	90.7	81.1	79.1	61.8	46.9	34.5	100	53.5
Cr	36.9	38.4	76.6	81.9	67.0	63.4	53.4	40.0	28.2	73.6	44.1
Co	3.93	4.32	15.5	15.1	13.6	13.4	10.3	5.58	4.46	13.8	6.40
Ni	11.0	11.3	38.6	40.5	32.4	31.5	26.4	11.7	9.39	39.4	12.4
<sup>143</sup> Nd/ <sup>144</sup> Nd ± 2σ	0.511758 ± 9	0.511749 ± 7	0.511752 ± 6	0.511736 ± 8	0.511748 ± 6	0.511745 ± 7	0.511755 ± 6	0.511719 ± 5	0.511788 ± 11	0.511767 ± 8	0.511769 ± 7
<sup>176</sup> Hf/ <sup>177</sup> Hf ± 2σ	0.282027 ± 11	0.282009 ± 7	0.282241 ± 9	0.282226 ± 6	0.282205 ± 6	0.282209 ± 6	0.282133 ± 7	0.281992 ± 11		0.282268 ± 7	0.281930 ± 6
<sup>208</sup> Pb/ <sup>204</sup> Pb ± 2σ	40.6213 ± 43	40.579 ± 39	40.3003 ± 18	40.3019 ± 25	40.2931 ± 34	40.2725 ± 16	40.1312 ± 28	40.8292 ± 23	40.8030 ± 26	39.9963 ± 28	42.4525 ± 27
<sup>207</sup> Pb/ <sup>204</sup> Pb ± 2σ	15.9733 ± 14	15.9644 ± 13	15.8991 ± 6	15.9013 ± 5	15.904 ± 7	15.9040 ± 6	15.8984 ± 10	15.9684 ± 9	15.9548 ± 9	15.873 ± 13	16.1109 ± 9
<sup>206</sup> Pb/ <sup>204</sup> Pb ± 2σ	20.7277 ± 14	20.6899 ± 18	20.0301 ± 7	20.0574 ± 9	20.0912 ± 8	20.0756 ± 7	20.0387 ± 11	20.6767 ± 9	20.5845 ± 10	19.7765 ± 10	22.1618 ± 10