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Abstract: The Nd and Hf isotopic compositions of most crustal and mantle rocks correlate to form the "Terrestrial Array". However, it is now well established that whereas coarse detrital sediments follow this trend, fine-grained oceanic sediments have high Hf ratios relative to their Nd isotopic ratios. It remains uncertain whether this "decoupling" of the two isotopic systems only occurs in the oceanic environment or if it is induced by sedimentary processes in continental settings. Here, we focus on the Hf and Nd isotopic compositions of sediments in large rivers in order to constrain the behavior of the two isotopic systems during erosion and sediment transport from continent to ocean. We report major and trace element concentrations together with Nd and Hf isotopic compositions of bedloads, suspended loads and river banks from the Ganges River and its tributaries draining the Himalayan Range i.e. the Karnali, the Narayani, the Kosi and the Marsyandi Rivers. Our sample set includes sediments sampled within the Himalayan Range in Nepal, at the Himalayan mountain front, and also downstream on the floodplain and at the outflow of the Ganges in Bangladesh. We show that hydrodynamic sorting of minerals explains the entire Hf isotopic range, i.e. more than 10 ϵ_{Hf} units, observed in the river sediments but does not affect the Nd isotopic composition. Bedloads and bank sediments have systematically lower ϵ_{Hf} values than suspended loads sampled at the same location. Coarse-grained sediments lie below or on the Terrestrial Array in an ϵ_{Hf} vs. ϵ_{Nd} diagram. In contrast, fine-grained sediments, including most of the suspended loads, deviate from the Terrestrial Array toward higher ϵ_{Hf} relative to their ϵ_{Nd} , as is the case for oceanic terrigenous clays. We explain the observed Nd-Hf decoupling by mineralogical sorting processes that enrich bottom sediments in coarse and dense minerals, including unradiogenic zircons, while surface sediments are enriched in fine material with radiogenic Hf signatures. We demonstrate that Nd-Hf isotopic decoupling increases with sediment transport in the floodplain to reach its maximum at the river mouth. The Nd-Hf isotopic decoupling observed in worldwide oceanic clays and river sediments is likely to have the same origin. Finally, we estimated the Nd-Hf isotopic composition of the present-day mantle if oceanic sediments had never been subducted and conclude that the addition of oceanic sediments with their anomalous Nd-Hf isotopic compositions has slowly shifted the composition of the Earth's mantle towards more radiogenic Hf values.

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Continental sedimentary processes decouple Nd and Hf isotopes

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35 **Abstract**

36 The Nd and Hf isotopic compositions of most crustal and mantle rocks correlate to form
37 the “Terrestrial Array”. However, it is now well established that whereas coarse detrital
38 sediments follow this trend, fine-grained oceanic sediments have high Hf ratios relative
39 to their Nd isotopic ratios. It remains uncertain whether this “decoupling” of the two
40 isotopic systems only occurs in the oceanic environment or if it is induced by
41 sedimentary processes in continental settings. Here, we focus on the Hf and Nd isotopic
42 compositions of sediments in large rivers in order to constrain the behavior of the two
43 isotopic systems during erosion and sediment transport from continent to ocean.

44 We report major and trace element concentrations together with Nd and Hf isotopic
45 compositions of bedloads, suspended loads and river banks from the Ganges River and
46 its tributaries draining the Himalayan Range i.e. the Karnali, the Narayani, the Kosi and
47 the Marsyandi Rivers. Our sample set includes sediments sampled within the Himalayan
48 Range in Nepal, at the Himalayan mountain front, and also downstream on the
49 floodplain and at the outflow of the Ganges in Bangladesh. We show that hydrodynamic
50 sorting of minerals explains the entire Hf isotopic range, i.e. more than 10 ϵ_{Hf} units,
51 observed in the river sediments but does not affect the Nd isotopic composition.

52 Bedloads and bank sediments have systematically lower ϵ_{Hf} values than suspended loads
53 sampled at the same location. Coarse-grained sediments lie below or on the Terrestrial
54 Array in an ϵ_{Hf} vs. ϵ_{Nd} diagram. In contrast, fine-grained sediments, including most of the
55 suspended loads, deviate from the Terrestrial Array toward higher ϵ_{Hf} relative to their
56 ϵ_{Nd} , as is the case for oceanic terrigenous clays. We explain the observed Nd-Hf
57 decoupling by mineralogical sorting processes that enrich bottom sediments in coarse
58 and dense minerals, including unradiogenic zircons, while surface sediments are

59 enriched in fine material with radiogenic Hf signatures. We demonstrate that Nd-Hf
60 isotopic decoupling increases with sediment transport in the floodplain to reach its
61 maximum at the river mouth. The Nd-Hf isotopic decoupling observed in worldwide
62 oceanic clays and river sediments is likely to have the same origin. Finally, we estimated
63 the Nd-Hf isotopic composition of the present-day mantle if oceanic sediments had
64 never been subducted and conclude that the addition of oceanic sediments with their
65 anomalous Nd-Hf isotopic compositions has slowly shifted the composition of the
66 Earth's mantle towards more radiogenic Hf values.

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68 **Keywords:** Nd, Hf, river sediments, decoupling of isotopic systems, mineralogical
69 sorting

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83 **1. Introduction**

84 The present-day Nd and Hf isotopic compositions of most crustal and mantle-
85 derived rocks correlate and form the “Terrestrial Array” (Vervoort et al., 1999; Vervoort
86 et al., 2011) due to similar fractionation of Sm/Nd and Lu/Hf during magmatic
87 processes. However, it is also recognized that some oceanic sediments, in particular
88 oceanic ferromanganese crusts and terrigenous clays, deviate significantly from the
89 Terrestrial Array towards higher ϵ_{Hf} values relative to their ϵ_{Nd} values (e.g. Albarède et
90 al., 1998; Vervoort et al., 1999; van de Fliedrt et al., 2004; Chauvel et al., 2008; Bayon et
91 al., 2009; Vervoort et al., 2011). The exact cause of this shift in Nd-Hf isotopic
92 compositions is still debated (e.g. Bau and Koschinsky, 2006; van de Fliedrt et al., 2007;
93 Vervoort et al., 2011). Patchett et al. (1984), Chauvel et al. (2008), Carpentier et al.
94 (2009) and Vervoort et al. (2011) suggest that the decoupling in terrigenous oceanic
95 sediments is due to sequestration on continents of zircons with low ϵ_{Hf} : the so-called
96 “zircon effect” e.g., a process that transports material with elevated Lu/Hf and Hf
97 isotopic ratios to the ocean basins. Authors who mainly concentrated on seawater and
98 ferromanganese crusts have more diverse interpretations. For example, Pettke et al.
99 (2002) and Rickli et al. (2010) evaluated the input of eolian dusts with high ϵ_{Hf} values
100 while Bayon et al. (2006), van de Fliedrt et al. (2007), Bayon et al. (2009), Godfrey et al.
101 (2009), Chen et al. (2011), Vervoort et al., (2011) and Rickli et al. (2013) suggested that
102 the decoupling could be due to preferential release of radiogenic Hf during chemical
103 weathering of phases with high Lu/Hf such as apatite.

104 Almost all studies suggest that the Nd and Hf isotopes are decoupled on
105 continents and inherited by the oceanic sediments. However, only few studies have
106 focused on continental sediments (e.g. Bayon et al., 2006; Rickli et al., 2013) and Nd-Hf
107 isotopic systematics of river sediments have never been thoroughly investigated. A

108 proper understanding of these processes is required if Nd-Hf isotopes are to be used as
109 provenance and weathering proxies and also, at a more global scale, to determine the
110 Nd-Hf isotopic composition of the continental crust and mantle. We currently do not
111 know whether zircon-rich sediments largely remain on the continents or are
112 transported into ocean basins. In addition, if the composition of sediment recycled into
113 the mantle in subduction zones differs from average continental crust, how does this
114 decoupling of the two isotopic systems affect the long-term evolution of the mantle?

115 The aim of this paper is to accurately document the Nd-Hf isotopic compositions
116 of sediments transported by large rivers to better understand the behavior of the two
117 isotopic systems during erosion and sediment transport from land to sea. To accomplish
118 this, we use a comprehensive sample set including bedloads, bank deposits and
119 suspended loads at the Himalayan front and floodplain of the Ganges, Karnali, Narayani
120 and Kosi Rivers, along the Marsyandi River in the Himalayan Range in Nepal, and at the
121 mouths of small tributaries draining single geological units.

122 The Ganges River and its tributaries constitute one of the largest fluvial systems
123 in the world (Lupker et al., 2011). The immense size of this fluvial network, together
124 with its importance in the world sedimentation system, provide the opportunity to
125 study global Nd-Hf isotopic systematics of river sediments. Sediments from the Ganges
126 and its major tributaries have been widely studied, in particular (a) in provenance
127 studies (Galy and France-Lanord, 2001; Garzanti et al., 2007; Tripathi et al., 2007; Singh
128 et al., 2008; Singh, 2009; Singh, 2010), (b) to determine the effects of chemical
129 weathering (Galy and France-Lanord, 1999; Lupker et al., 2012), (c) to characterize
130 mineralogical variations associated to hydraulic sorting (Garzanti et al., 2010; Garzanti
131 et al., 2011), (d) to quantify organic carbon transfer (Galy et al., 2007; Galy et al., 2008)
132 and (e) to estimate global sediment flux budgets (Galy and France-Lanord, 2001; Lupker

133 et al., 2011). Using these studies and our new results, we investigated the effects of
134 sediment provenance and mineral sorting during sediment transport on the trace-
135 element concentrations and Nd-Hf isotopic compositions of these river sediments.

136

137 **2. The Ganges fluvial system**

138 The Ganges fluvial system is one of the main conveyors of material eroded from
139 the Himalayan Range, delivering ca. 400 million tons of sediments per year (Lupker et
140 al., 2011) to the ocean in the Bay of Bengal. Together with the Brahmaputra River, the
141 Ganga ranks among the three largest fluvial systems in terms of annual sediment load
142 delivered into the ocean (Milliman, 2001). The sediments accumulate in the Bengal fan,
143 by far the largest fan in the world with its volume of $12.5 \times 10^6 \text{ km}^3$ and a sediment mass
144 of ca. 3×10^{16} tons (Curry, 1994; Curry et al., 2003).

145 The Ganges and its major tributaries drain an area of about 1 million km^2 that
146 stretches over 2500 km from its sources in the High Himalayan Range to its delta in
147 Bangladesh (Figure 1). The water discharge of the Ganges is strongly controlled by the
148 monsoon from June to September, which accounts for about 75% of its annual flow in
149 Bangladesh (Singh et al., 2007). The Ganges basin (Figure 1) includes several major
150 tributaries, the Yamuna, Karnali, Narayani and Kosi Rivers, which drain the southern
151 flank of the Himalayan Range in the north and the Chambal River, which drains part of
152 the Indian shield and the Deccan Traps basalts in the south. Le Fort (1975) divided the
153 Ganges drainage basin in the Himalayan Range, into four geological units: (1) the
154 Tethyan Sedimentary Series, which consists of weakly metamorphosed Cambrian to
155 Eocene carbonates and clastic sedimentary rocks (dark grey in Figure 1); (2) the High
156 Himalayan Crystalline units mostly composed of strongly metamorphosed
157 Neoproterozoic to Ordovician gneiss, migmatites, and marbles (light grey in Figure 1);

158 (3) the Lesser Himalaya units of weakly metamorphosed Proterozoic sedimentary rocks
159 (white in Figure 1); and (4) the Neogene to Quaternary Siwalik molasses which are
160 floodplain deposits of recycled erosion products from the Himalayan Range (orange in
161 Figure 1). France-Lanord et al. (1993) and Galy and France-Lanord (2001) showed that
162 more than two thirds of the sediments transported by the Ganges River derived from the
163 High Himalayan Crystalline units, with the remainder coming from the Lesser Himalaya
164 units.

165 The mineralogy of Ganges sediments includes both inherited and neofomed
166 mineral phases and is controlled by hydrodynamic processes during fluvial transport
167 (see Galy et al., 2008; Garzanti et al., 2010; Garzanti et al., 2011). Supplementary Table A
168 reports estimates of Garzanti et al. (2010, 2011) of the average mineral proportions of
169 bedloads and suspended loads sampled in the Ganges in Bangladesh. According to these
170 studies, bedloads contain higher proportions of quartz (40 to 60%) and heavy minerals
171 (up to 15%) than suspended loads (27 to 55% quartz and 3 to 6% heavy minerals).
172 Heavy minerals include amphibole, epidote, garnet and many accessories such as
173 titanite, zircon, monazite, allanite, apatite and tourmaline (Supplementary Table A). By
174 contrast, suspended load sediments tend to be enriched in phyllosilicates such as clays
175 (8 to 26%) and micas (9 to 30%).

176

177 **3. Sediment Sampling and analytical procedure**

178 **3.1. Sediment Sampling**

179 River sediments analyzed in this study were sampled over several years of field
180 campaigns, in Nepal, India and Bangladesh. Some of these samples have already been
181 analyzed for major elements (Lupker et al., 2012), Re-Os isotopes (Pierson-Wickmann et

182 al., 2000), mineralogical proportions (Garzanti et al., 2007; 2010; 2011) and C_{org}
183 isotopes (Galy et al., 2008).

184 To document the Nd-Hf isotopic variability of river sediments at a large scale, i.e.
185 during their transport from the mountain range to the delta, we analyzed a set of
186 samples from the Ganges and its major tributaries (Karnali, Narayani and Kosi Rivers)
187 collected at the front of the Himalayan Range and in the floodplain (Figure 1). Because
188 more than 95% of the suspended load is transported during the monsoon season (Islam
189 et al., 1999), most samples were collected between July and September from 1993 to
190 2010. We also studied the isotopic variability of the sediments at a smaller scale using
191 10 samples collected in the Himalayas along the Marsyandi River, from its source at ca.
192 5000 m altitude to its confluence with the Trisuli River in Nepal at an altitude of ca. 250
193 m (pink star in Figure 1). The precise locations of these samples, together with a
194 detailed description of the Marsyandi Basin, can be found in Attal and Lavé (2006) and
195 Garzanti et al. (2007) while GPS derived coordinates are provided in Supplementary File
196 B. Finally, we analyzed sediments from small Nepalese rivers draining monolithologic
197 basins to characterize the chemical and isotopic compositions of the three main
198 Himalayan geological units i.e. the Lesser Himalaya, the High Himalayan Crystalline and
199 the Tethyan Sedimentary Series (white, light grey and dark grey stars in Figure 1).

200 To study the effects of mineralogical sorting on the eroded material transported
201 by the Ganges, we analyzed different types of river sediment at each sampling site.
202 Wherever possible, the suspended load was sampled at the surface and at different
203 depths in the water column, and additional samples were obtained from dredged
204 bedload sediments and bank sediments. Sampling techniques for suspended load,
205 bedload and bank sediment are provided in Galy et al. (2008) and Lupker et al. (2011).

206

207 **3.2. Chemical analyses**

208 **3.2.1. Major element contents**

209 Major element contents were measured in the sediments from the Marsyandi
210 River and from the Nepalese rivers draining single geological units. Major element
211 concentrations in all the other studied samples were published by Lupker et al. (2012)
212 (See Supplementary Table B). After freeze-drying and grinding with an agate mortar to
213 ensure homogeneity, about 100 mg of sediments were fused using lithium metaborate
214 to measure major element concentrations by ICP-AES at the SARM in Nancy (France)
215 according to the technique described by Govindaraju and Mevelle (1987) and Carignan
216 et al. (2007).

217

218 **3.2.2. Trace element concentrations and Nd-Hf isotopes**

219 **3.2.2.a. Sample preparation and dissolution**

220 The samples were further ground in agate bowls using a planetary ball mill to
221 insure fine grain size. About 50 to 100 mg of powder were initially treated with 5 mL of
222 14N HNO₃ on a hotplate for 2 days. After evaporation, the samples were digested in a
223 10:1 mixture of 5 mL HF and 0.5 mL HClO₄ using teflon containers placed in steel PARR
224 bombs for 6 weeks at 140°C or one week at 200°C. The digestion time and temperature
225 needed to completely dissolve resistant minerals such as zircons were previously
226 estimated by several tests comparing (1) the trace element content of river sediments
227 (suspended load, bedload and bank deposit from the Ganges) being digested at different
228 temperatures during 1, 3 and 6 weeks with (2) the trace element content of the same
229 samples being fused with lithium metaborate, a technique that ensures total digestion of
230 all resistant minerals. Such tests indicate that sediments must be digested during at least

231 6 weeks at 140°C or one week at 200°C to ensure total dissolution of zircons and other
232 resistant minerals. After complete dissolution, two aliquots were taken from each
233 sample: one to measure trace element concentrations and the other to isolate Nd and Hf
234 for isotopic measurements.

235

236 **3.2.2.b. Trace element concentrations**

237 Trace element concentrations were measured using an Agilent 7500ce ICP-MS in
238 Grenoble (France) following the procedure described by Chauvel et al. (2011).
239 Measurements were performed on samples diluted 5000 times in a solution of 2% HNO₃
240 with traces of HF. They were corrected for oxide interference and ICP-MS analytical drift
241 using a multispiked of Be, Ge, In, Tm and Bi. Repeated measurements of BR or BR-24 rock
242 standards were used as an external calibration to calculate trace element concentrations
243 in all samples. The validity and the reproducibility of our results are assessed by
244 repeated measurements of international rock standards as unknown samples, including
245 sedimentary materials such as JSd-2, and the measurements of several complete
246 duplicates (see Supplementary Tables B and C).

247

248 **3.2.2.c. Nd-Hf isotopic compositions**

249 Hf and Nd were isolated using ion chromatography (Chauvel et al., 2011). Total
250 procedural blanks reached on average 94 pg (n=6) for Nd and 47 pg (n=11) for Hf and
251 are negligible compared to the amount of Nd and Hf isolated in all samples (on average
252 1600 ng for Nd and 600 ng for Hf). Neodymium and Hf isotopic compositions were
253 analyzed on a Nu Plasma HR MC-ICP-MS at the ENS Lyon (France) and were corrected
254 for mass fractionation bias using $^{146}\text{Nd}/^{144}\text{Nd}=0.7219$ and $^{179}\text{Hf}/^{177}\text{Hf}=0.7325$,

255 respectively. The average measured values for the Ames-Rennes Nd and Ames-Grenoble
256 Hf reference standards run every two or three samples are $^{143}\text{Nd}/^{144}\text{Nd} = 0.511959 \pm 8$
257 (2σ , $n = 59$) and $^{176}\text{Hf}/^{177}\text{Hf} = 0.282163 \pm 10$ (2σ , $n = 99$), respectively. MC-ICP-MS
258 analytical drift was corrected using the recommended values for these two standards
259 ($^{143}\text{Nd}/^{144}\text{Nd} = 0.511961 \pm 13$; Chauvel and Blichert-Toft (2001) and $^{176}\text{Hf}/^{177}\text{Hf} =$
260 0.282160 ± 12 ; Chauvel et al. (2011)). The reproducibility of our results estimated using
261 complete duplicate analyses is usually better than 0.5 ϵ units for Nd and Hf isotopic
262 compositions (see Table 1).

263

264 **4. Results**

265 **4.1. Major and Trace element concentrations**

266 Major element contents are presented in Supplementary Table B. For the purpose of this
267 study, we focus on the SiO_2 and Al_2O_3 contents of the studied bedloads, bank deposits
268 and suspended loads, as shown in Figure 2a. Lupker et al. (2011, 2012) describe the
269 variations of other major elements. In general, SiO_2 and Al_2O_3 contents are strongly
270 correlated with sediment grain-size (Lupker et al., 2011). Bedloads and bank sediments
271 are enriched in coarse-grained quartz and have high SiO_2 contents (ca. 70-80 wt.%), and
272 relatively low Al_2O_3 (ca. 5-12 wt.%), Fe_2O_3 (ca. 2-5 wt.%), K_2O (ca. 1-3 wt.%), MgO (ca. 1-
273 3 wt.%) and TiO_2 (ca. 0.1-0.6 wt.%). Suspended loads have lower SiO_2 (ca. 50-70 wt.%)
274 and tend to have higher contents of elements hosted by fine phyllosilicates such as Al_2O_3
275 (ca. 10-18 wt.%), Fe_2O_3 (ca. 4-8 wt.%), K_2O (ca. 3-5 wt.%), MgO (ca. 2-5 wt.%) and TiO_2
276 (ca. 0.5-1 wt.%). Due to this chemical differentiation, $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios can be used as a
277 reliable grain-size proxy for detrital sediments: Figure 2b shows that bedloads and bank
278 deposits have lower $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios than suspended loads. Moreover, Lupker et al.

279 (2011) showed that $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios of suspended loads are very well correlated with
280 their sampling depth in the water column. This feature, not shown in Figure 2, accounts
281 for the variability of $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios in suspended loads: surface suspended loads have
282 higher $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios than those sampled at greater depths. It can also be seen in
283 Figure 2 that some of the bedloads and bank deposits have surprisingly high $\text{Al}_2\text{O}_3/\text{SiO}_2$
284 ratios. These samples were recovered during the dry season when weak water flow
285 favored deposition of Al_2O_3 -rich phyllosilicates in bedloads and bank sediments.

286 Trace-element concentrations are presented in the Supplementary Table B.
287 Spidergrams normalized to the average upper continental crust composition of Rudnick
288 and Gao (2003) are shown in Figure 3 for sediments from the Nepalese rivers draining
289 exclusively the Tethyan Sedimentary Series, the High Himalayan Crystalline and the
290 Lesser Himalaya units (Figure 3a-c) and for the Marsyandi, Ganges, Narayani, Karnali
291 and Kosi Rivers (Figure 3d-h).

292 In the Himalayan Range, sediments from small Nepalese rivers draining single
293 sources display variable trace element distributions (Figure 3a-c). They are
294 systematically depleted in transition elements and mobile elements such as Ba and Sr.
295 Enrichment factors are generally higher for the Tethyan Sedimentary Series than for the
296 High Himalayan Crystalline and the Lesser Himalaya units. The trace-element
297 characteristics can be quantified using element concentrations and ratios normalized to
298 the average upper continental crust (e.g. $(\text{Th}/\text{U})_{\text{UCC}}$). Sediments from Tethyan
299 Sedimentary Series show clear enrichments in Cs, Rb, Li and As, very high $(\text{Th}/\text{U})_{\text{UCC}}$
300 ratios (2.0 ± 0.6 (1σ) on average) and rather flat REE patterns (average $(\text{La}/\text{Yb})_{\text{UCC}}$ of
301 1.2 ± 0.3 (1σ)). By contrast, the REE patterns of sediments eroded from High Himalayan
302 Crystalline units are characterized by significant enrichments in heavy REE (average

303 (La/Yb)_{UCC} of 0.4±0.2 (1σ). Lesser Himalaya sediments show much more variability
304 with (La/Yb)_{UCC} ratios ranging from 0.7 to 1.4.

305 Trace-element concentrations measured in sediments collected along the high-
306 altitude Marsyandi river are more variable than those sampled in rivers on the
307 floodplain (see Figure 3d-h). When compared to the average upper continental crust
308 composition, most are enriched in Cs, Rb and depleted in Ba, Sr and transition elements
309 (Sc, V, Cr, Co, Ni, Cu, Zn and As). Enrichment factors for Zr-Hf range from 0.3 to 1.5 and
310 from 0.8 to 4.4 for Th-U. Their REE patterns display a small enrichment in heavy REE, as
311 shown by their average (La/Yb)_{UCC} ratio of 0.7±0.3 (1σ).

312 In the floodplain, trace-element patterns for sediments from the Ganges and its
313 major tributaries draining the Himalayan Range i.e. the Narayani, the Karnali and the
314 Kosi Rivers resemble each other (Figure 3e-h). No systematic downstream variation in
315 their composition occurs and trace-element distributions are globally comparable to the
316 composition of average upper continental crust, except for the transition elements and
317 some mobile elements (Ba, Sr) which are always depleted. REE patterns are generally
318 flat with an average (La/Yb)_{UCC} ratio of 0.8±0.1 (1σ) for the Ganges sediments, 0.9±0.1
319 (1σ) for the Narayani sediments, 0.9±0.2 (1σ) for the Karnali sediments and 0.9±0.1 (1σ)
320 for the Kosi sediments. There is no significant REE fractionation between bedload,
321 suspended load and bank deposits (average (La/Yb)_{UCC} of 0.9±0.2 (1σ) for all of the
322 bedloads and bank sediments and 0.9±0.1 (1σ) for all of the suspended loads). In
323 general, bedloads and bank sediments are characterized by a positive anomaly in Zr-Hf
324 and are more depleted than suspended loads in transition elements and mobile
325 elements such as Cs, Rb and Ba. In the Ganges River, bedloads and bank sediments show
326 significant enrichments in all REE and positive Th-U anomalies (Figure 3e). These

327 features are not observed in the Narayani, the Karnali and the Kosi Rivers for which the
328 chemical compositions of bedloads, bank deposits and suspended loads are similar.

329

330 **4.2. Nd-Hf isotopic compositions**

331 Neodymium and Hf isotopic compositions measured in this study are given in
332 Table 1. Sediments sampled in the Himalayan Range from rivers draining single
333 geological units and from the Marsyandi River are shown in a ϵ_{Hf} vs. ϵ_{Nd} diagram in
334 Figure 4 together with worldwide isotopic compositions of oceanic sediments.
335 Sediments from the floodplain of the Ganges and its major tributaries are shown in
336 Figure 5.

337 In the Himalayan Range (Figure 4), Nd-Hf isotopes of sediments from the Lesser
338 Himalaya units display significantly less radiogenic values ($-40.3 < \epsilon_{\text{Hf}} < -28.8$ and -25.1
339 $< \epsilon_{\text{Nd}} < -20.1$) than those from the High Himalayan Crystalline units ($-22.2 < \epsilon_{\text{Hf}} < -12.7$
340 and $-17.7 < \epsilon_{\text{Nd}} < -12.9$) and the Tethyan Sedimentary Series ($-23.7 < \epsilon_{\text{Hf}} < -19.5$ and $-$
341 $18.3 < \epsilon_{\text{Nd}} < -16.6$). The ϵ_{Nd} values are comparable to Nd isotopic compositions published
342 for bedrocks from the Lesser Himalaya, the High Himalayan Crystalline and the Tethyan
343 Sedimentary Series (see the histogram in Figure 4 and references therein).

344 Neodymium and Hf isotopic compositions of bedloads sampled all along the
345 Marsyandi River (Figure 4) are much less variable ($-25.5 < \epsilon_{\text{Hf}} < -19.9$ and $-18.0 < \epsilon_{\text{Nd}} < -$
346 15.4) and resemble those of the High Himalayan Crystalline and the Tethyan
347 Sedimentary Series. There is no clear downstream variation in ϵ_{Hf} and ϵ_{Nd} values (see
348 Figure 6; Table 1).

349 In the floodplain (Figure 5), Nd-Hf isotopic compositions of sediments from the
350 Ganges River and its major tributaries are similar. The Hf isotopic compositions are
351 nevertheless more variable ($-30.2 < \epsilon_{\text{Hf}} < -18.3$) than Nd isotopes ($-20.1 < \epsilon_{\text{Nd}} < -15.6$).

352 With the exception of few outliers, bedloads and bank sediments tend to lie below or on
353 the Terrestrial array while suspended loads, which have higher ϵ_{Hf} values, usually plot
354 above the array with compositions approaching those of worldwide oceanic terrigenous
355 clays. By contrast, suspended loads, bedloads and bank deposits cannot be distinguished
356 using their Nd isotopic compositions.

357

358 **5. Discussion**

359 **5.1. Source isotopic variability**

360 The Nd and Hf isotopic compositions of sediments along a river course are
361 influenced by contributions of materials with different isotopic compositions that are
362 supplied by tributaries. These source effects need to be evaluated before we can
363 consider any other control on the geochemistry of river sediments.

364 REE barely fractionate during weathering and have been widely used for
365 studying the provenance of detrital sediments. More specifically, ratios such as La/Yb, or
366 Nd isotopic compositions of weathered material, are considered to represent the
367 compositions of source rocks (Goldstein and Jacobsen, 1988; Condie, 1991; Taylor and
368 McLennan, 1995). We evaluate below how these proxies vary in the Himalayan river
369 sediments.

370

371 **5.1.1. The Marsyandi River in the Himalayan Range**

372 In the Himalayan Range, the Marsyandi River has its source at ca. 5000m altitude
373 in the Tethyan Sedimentary Series, then flows down through High Himalayan Crystalline
374 and Lesser Himalaya units to the floodplain at ca. 250 m altitude (Figure 6). Samples
375 recovered upstream, where the river drains only the Tethyan Sedimentary Series

376 (samples NAG 22 and MAR-50), are characterized by an average $(La/Yb)_{UCC}$ ratio of
377 1.2 ± 0.3 (1σ) and a ϵ_{Nd} value of -16.8 ± 0.3 (1σ) (Table 1).

378 Downstream, when the river passes through the High Himalayan Crystalline units
379 (samples MAR-55 and MAR-58), the average $(La/Yb)_{UCC}$ ratio drops to 0.7 ± 0.4 (1σ)
380 while average ϵ_{Nd} values are comparable to values obtained upstream ($\epsilon_{Nd} = -16.2 \pm 0.1$
381 (1σ)). The REE patterns of sediments are thus clearly affected by materials eroded from
382 the High Himalayan Crystalline units, which have an average $(La/Yb)_{UCC}$ ratio of 0.4 ± 0.2
383 (1σ) (see section 4.1). Because the Nd isotopic signature of the Tethyan Sedimentary
384 Series is similar to that of the High Himalayan Crystalline units (see the histogram in
385 Figure 4 and references therein), there is no significant change in the average ϵ_{Nd} values
386 of the sediments.

387 Farther downstream, sediments sampled where the river passes through the
388 Lesser Himalaya units (all MAR samples downstream of MAR-58) are characterized by
389 an average $(La/Yb)_{UCC}$ ratio of 0.6 ± 0.3 (1σ) and an ϵ_{Nd} value of -16.7 ± 1.1 (1σ). This value
390 is remarkably similar to that measured upstream even though the Lesser Himalaya units
391 have a significantly less radiogenic Nd isotopic signature (see the histogram in Figure 4
392 and references therein). This shows that the Lesser Himalaya units contribute little to
393 the final sediment load of the Marsyandi River, as already suggested by Garzanti et al.
394 (2007). Using petrographic and mineralogical data, Garzanti et al. (2007) suggest
395 contributions of $13 \pm 5\%$, $68 \pm 3\%$ and $20 \pm 2\%$ from the Tethyan Sedimentary Series, the
396 High Himalayan Crystalline and the Lesser Himalayan units, respectively. Assuming
397 comparable Nd concentrations for sediments from the three main sources, our mixing
398 calculations show that addition of 20% Lesser Himalayan sediments with an ϵ_{Nd} of -23.2,
399 to sediments derived from the other two sources ($\epsilon_{Nd} \approx -16.5$) only lowers the isotopic
400 composition of the mixture by 1.3 ϵ_{Nd} unit, a value consistent with our measurements.

401 In summary, despite the fact that the Marsyandi River crosses several geological
402 units with distinct isotopic signatures ($-25 < \epsilon_{Nd} < -13$), its sediments have rather constant
403 ϵ_{Nd} values of -16.6 ± 2.1 (1σ) on average. The variability of Nd isotopic compositions due
404 to source effects is limited (i.e. about 3 ϵ_{Nd} units) and the sediments are effectively
405 homogenized within the Himalayan Range before they enter the floodplain.

406

407 **5.1.2. The Ganges and its major tributaries in the floodplain**

408 Floodplain sediments of the Ganges and its major tributaries have remarkably
409 similar REE patterns with an average $(La/Yb)_{UCC}$ ratio of 0.88 (see section 4.1. and
410 Figure 3). As shown in Figure 5, their Nd isotopic compositions span a small range of
411 values (less than 5 ϵ_{Nd} units) similar to values obtained for the bedrocks from the High
412 Himalayan Crystalline units and the Tethyan Sedimentary Series (Figure 4). Nor is there
413 any systematic change in REE patterns and Nd isotopic compositions of river sediments
414 sampled at the Himalayan mountain front and farther downstream in the floodplain.
415 This indicates that no significant amount of new material is added along the course of
416 the Ganges, as already suggested in earlier studies using major and trace elements
417 (Tripathi et al., 2007; Singh et al., 2008; Singh, 2009; Lupker et al., 2012).

418 Sediments derived from erosion of the Indian shield and the Deccan Traps are
419 also added to the Ganges (see Figure 1) and one might expect that these terranes could
420 influence the Nd isotopic composition of sediments after their confluence. However,
421 Lupker et al. (2012) excluded significant contributions from these sources and
422 estimated the contribution of the southern tributaries to the entire Ganges sediment
423 load to be less than 4% in Bangladesh when our samples were recovered in 2004,
424 2007 and 2008. Our new ϵ_{Nd} values support this interpretation in that sediments
425 collected in Bangladesh have values similar to upstream samples.

426 Within the range of ϵ_{Nd} values obtained for the floodplain samples (-20 to -15),
427 the most negative values are likely due to higher contributions of sediments from the
428 Lesser Himalaya units depending on the sampling period and/or location. This could
429 notably be the case for the Kosi and Narayani sediments (green and purple dots in
430 Figure 5). For example, the ϵ_{Nd} values of the Narayani samples, which were all collected
431 at the same location in Nepal, vary from -19.5 to -17.3 depending on sampling time (July
432 2005, August 2007 and July 2010). This suggests that part of the Nd isotopic variability
433 in Figure 5 could be temporal and related to inefficient local homogenization of the
434 sediments due to changes of the river flow conditions throughout the hydrological cycle
435 or from a year to the other. Such a phenomenon was already advocated by Lupker et al.
436 (2012) for Ganges sediments sampled in Bangladesh and by Viers et al. (2008) for the
437 Amazon River.

438

439 **5.2. Effects of hydrodynamic sorting of minerals**

440 Sediments transported by rivers consist of particles and minerals with different
441 hydrodynamic properties that depend on their size, shape and density but are
442 independent of their primary or secondary origin. This results in “mineral sorting”
443 within the water column: coarse and dense grains are preferentially transported in
444 bedload by sliding and bouncing while fine and light grains are preferentially
445 transported in suspended load and at different depth in the water column depending on
446 their settling velocities. The most common coarse and dense grains usually consist of
447 minerals resistant to weathering processes such as quartz, zircon or monazite. In
448 contrast, the light and fine grains in the suspended load include primary mineral phases
449 such as mica as well as secondary minerals formed during weathering (clays). Because
450 chemical elements are present in different concentrations in minerals, mineral sorting

451 produces significant geochemical variability in river sediments (Singh and France-
452 Lanord, 2002; Garzanti et al., 2010; Bouchez et al., 2011; Garzanti et al., 2011; Lupker et
453 al., 2011). To assess its impact on the Nd-Hf isotopic systematics of the Himalayan river
454 sediments, we first briefly describe its influence on major and trace element
455 concentrations, then focus on the Nd and Hf budgets.

456

457 **5.2.1. Impact on the overall chemical compositions**

458 The influence of mineral sorting during sediment transport on the major and
459 trace element concentrations of sediments from the Himalayan rivers was studied in
460 detail by Garzanti et al. (2010, 2011) and Lupker et al. (2011, 2012). Our new
461 measurements are consistent with results obtained by these authors. As the factor
462 controlling the Al_2O_3 and SiO_2 contents is sediment grain size (Lupker et al., 2011),
463 $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio can be used as a reliable grain-size proxy: coarse-grained sediments i.e.
464 most of the bedloads and bank deposits, are enriched in SiO_2 -rich quartz while finer-
465 grained sediments, i.e. most of the suspended load, are enriched in Al_2O_3 -rich
466 phyllosilicates (see Figure 2).

467 Systematic changes of trace-element concentrations between bedloads, bank
468 sediments and suspended loads also occur (Figure 3). Suspended loads generally have
469 higher concentrations of transition and mobile elements due to their high proportion of
470 phyllosilicates, oxyhydroxides, soil particles and organic matter (Garzanti et al., 2011).
471 For the other trace elements, differences between bedload, bank sediment and
472 suspended load are more difficult to explain because they result from the combined
473 effects of several processes: (1) contribution of mineral phases such as zircon, rutile or
474 monazite that strongly control the budget of Zr-Hf, Nb-Ta and REE, respectively (Garçon
475 et al., 2011); (2) the proportions of mineral species that occur in different quantities in

476 bedloads, banks and suspended loads; and (3) dilution by minerals such as quartz that
477 are relatively poor in trace elements. The combination of the three processes explains
478 why the trace element patterns of bedloads and bank sediments are not systematically
479 different from those of suspended loads in Figures 3f-g-h even though it is clear that the
480 two types of sediments do not share similar mineralogical compositions (cf.
481 Supplementary Table A and Garzanti et al., 2010; 2011). Bedloads and bank sediments
482 from the Ganges River are the only ones with trace-element patterns distinct from the
483 suspended loads (Figure 3e). As most Ganges sediments were sampled in the
484 Bangladesh delta, where river hydrodynamics are relatively calm, segregation of
485 minerals within the water column is more efficient and the difference between heavy
486 mineral-rich bedloads and clay-rich suspended loads is clearer than in sediments
487 sampled upstream in the floodplain.

488 Heavy minerals play a key role in the chemical budget of sediments because they
489 host specific elements in extreme concentrations: this is the case of Zr and Hf in zircon
490 and LREE, Th and U in monazite and allanite (e.g. Götze and Lewis, 1994; Totten, 2007;
491 Garçon et al., 2011). Along the Marsyandi River, Garzanti et al. (2007) show that the
492 proportion of heavy minerals in bank sediments is highly variable, ranging between 1
493 and 31 wt.% in the 63-250 μm fraction. We thus suggest that in the Marsyandi bank
494 sediments, highly variable proportions of heavy minerals are responsible for the large
495 variability of trace element contents (Figure 3d). Unfortunately, we cannot establish a
496 direct link between the heavy mineral proportions estimated by Garzanti et al. (2007)
497 and our trace element data because only four of our samples were analyzed by Garzanti
498 et al. (2007), and because the heavy mineral proportions determined by these authors
499 were not obtained on bulk samples but on the 63-250 μm fraction.

500 In the floodplain sediments, Zr-Hf anomalies are good proxies to the relative
501 proportion of zircon and REE-bearing minerals. Most bedloads and bank sediments from
502 the Ganges and its major tributaries exhibit a positive Zr-Hf anomaly, suggesting excess
503 zircons, as is to be expected in coarse-grained sediments. However, the absence of
504 positive Zr-Hf anomalies in most suspended loads and in some bedloads and bank
505 sediments from the Narayani and Kosi Rivers does not imply that these sediments lack
506 zircon; it simply means that the relative proportion of zircon to REE-bearing minerals is
507 lower. Indeed, Garzanti et al. (2010, 2011) demonstrated that zircon controls almost the
508 entire Zr-Hf budget of the Ganges bedloads and about half the Zr-Hf budget of surface
509 suspended loads. They also demonstrated that monazite and allanite control the entire
510 REE budget, including that of Nd in the Ganges bedloads, whereas phyllosilicates,
511 together with titanite and monazite, are the carriers of REE in suspended loads. Our new
512 trace element data further support this interpretation.

513

514 **5.2.2. Nd-Hf isotopic decoupling**

515 As discussed in section 5.2.1, Nd and Hf in bedloads and suspended loads are
516 carried by different minerals whose proportions in the two types of sediments are
517 controlled by sorting during transport. Since these minerals do not all have the same
518 Sm/Nd and Lu/Hf ratios and formed millions of years ago, the Nd and Hf isotopic
519 compositions of bedloads and suspended loads should differ.

520 Figures 4 and 5, where ϵ_{Hf} is plotted as a function of ϵ_{Nd} , show that bedloads and
521 bank sediments plot on or slightly below the Terrestrial Array while suspended loads
522 plot above. Figure 7 plots ϵ_{Nd} and ϵ_{Hf} as a function of $\text{Al}_2\text{O}_3/\text{SiO}_2$ i.e. the grain size proxy.
523 No clear change of ϵ_{Nd} occurs as a function of the sediment grain-size, but the ϵ_{Hf} of
524 coarse-grained sediments with low $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios are systematically lower than those

525 of fine-grained sediments with high $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios. This is particularly true when the
526 bedloads and bank sediments were sampled at the same location as the suspended loads
527 (see the color bands linking related samples in Figure 7). Given that source variability is
528 limited in these sediments (see section 5.1), our observations suggest that mineral
529 sorting during transport has a significant effect on Hf isotopic compositions but does not
530 change the Nd isotopic signature. Nd and Hf isotopes are thus decoupled in fine-grained
531 sediments with high $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios. This is confirmed by the remarkable correlation
532 between $\Delta\varepsilon_{\text{Hf}}$, a measure of the vertical ε_{Hf} deviation from the Terrestrial Array, and
533 $\text{Al}_2\text{O}_3/\text{SiO}_2$, as shown in Figure 8. With few exceptions, coarse-grained sediments lie
534 below or on the Terrestrial Array while $\Delta\varepsilon_{\text{Hf}}$ increases as grain size decreases in all other
535 sediments. We conclude that the entire Hf isotopic variability observed in the Himalayan
536 river sediments (more than 10 ε_{Hf} units) relates to changes in sediment grain size, and is
537 therefore due to mineral sorting during sediment transport.

538 Similar decoupling of Nd and Hf isotopes by mineral sorting has already been
539 reported in detrital oceanic materials (see Figure 4 and 5; Patchett et al., 1984; Chauvel
540 et al., 2008; Bayon et al., 2009; Carpentier et al., 2009; Vervoort et al., 2011) but, to our
541 knowledge, it has never before been demonstrated for modern river sediments on
542 continents. We believe that in both environments, sample mineralogy controls the
543 fractionation of Hf from Nd isotopes as described by Patchett et al. (1984). Zircon hosts
544 most of the Hf in river sediment and has very low Lu/Hf ratios (see Kinny and Maas,
545 2003 for a review) compared to any other mineral. Its Hf isotopic composition is
546 therefore very close to the initial ratio of the material from which it crystallized, and
547 always significantly less radiogenic than the Hf isotopic composition of all other crustal
548 minerals. Because coarse-grained sediments are zircon-rich, they inherit the
549 unradiogenic Hf isotopic composition of zircon. By contrast, the Hf isotopic composition

550 of fine-grained sediment varies depending on the relative proportion of zircon to other,
551 more radiogenic minerals with high Lu/Hf ratios. These minerals include primary
552 minerals formed during crystallization of the source rocks (i.e. muscovite, biotite) and
553 secondary minerals formed through alteration and weathering (i.e. clays). Hence, we do
554 not exclude that additional fractionation is introduced during differential chemical
555 weathering of minerals with high Lu/Hf as suggested by several authors (Bayon et al.,
556 2006; Godfrey et al., 2009; Chen et al., 2011; Vervoort et al., 2011, Rickli et al., 2013). Of
557 course, the amplitude of the Hf isotopic fractionation also depends on the age of the
558 crustal protolith since time is necessary to produce ^{176}Hf as a decay product of ^{176}Lu .

559 In contrast to Hf isotopes, mineral sorting appears to have minimal effect on Nd
560 isotopes because the minerals that host Nd in bedloads, suspended loads and bank
561 sediments (i.e. phyllosilicates, monazite, etc) have the same Sm/Nd ratio as their
562 protolith. As a consequence, all sediments have the same Nd isotopic composition i.e.
563 that of their source rocks. We demonstrated that this is clearly not the case for the Hf
564 isotopic composition of sediments which strongly depends on the sample mineralogy;
565 most sediments do not have the same Hf isotopic signature as their source rocks. Only
566 coarse-grained sediments, rich in zircons, have Hf isotopes that can be used to estimate
567 the composition of their continental sources provided that the age of the zircon
568 population is known (Harrison et al., 2005; Kemp et al., 2007; Garçon et al., 2011).

569

570 **5.3. Where does decoupling of Nd and Hf isotopes occur on** 571 **continents?**

572 Mineral sorting processes depend strongly on river hydrodynamics. Thus, the
573 chemical and isotopic compositions of river sediments should vary as a function of
574 sampling site, and values in the steep Himalayan Range should differ from those in the

575 floodplain. The extent of Nd-Hf isotopic decoupling in fine-grained sediments should
576 also vary depending on the degree of mineral sorting.

577 In the Himalayan Range, ϵ_{Nd} values in the Marsyandi sediments vary within a
578 small range between -18.0 and -15.4 (Figure 6) and are not significantly affected by
579 source effects or mineralogical sorting. The same applies to ϵ_{Hf} values of bedloads, even
580 though they are more variable (-25.5 to -19.9). The ϵ_{Hf} variability does not correlate with
581 the Nd isotopic compositions of sediments, and is not related to the geological units
582 through which the river flows (Figure 6). Because the modern river system does not
583 contain long-term storage sites (Attal and Lavé, 2006), the data suggests that the Hf
584 isotopic composition of the Marsyandi bedloads is controlled by local changes in the
585 river hydrodynamics that influence the proportions of heavy minerals (see section 5.2.1.
586 and Garzanti et al., 2007). We could not measure the isotopic compositions of suspended
587 loads in the Marsyandi River and we cannot discuss how different they are from the
588 bedloads. However, our bedload data alone suggest that the Hf isotopic variability of
589 sediments sampled in the mountain range is mainly controlled by local hydrodynamic
590 conditions.

591 The situation is very different, however, on the Ganges floodplain. Figure 9 shows
592 the variations of Nd and Hf isotopic compositions of bedloads, suspended loads and
593 bank sediments from the Ganges and its major tributaries as a function of sample
594 locations. While ϵ_{Nd} values are remarkably constant at about -17.6 ± 1 (1σ) and do not
595 change downstream, ϵ_{Hf} values display significant variations. As mentioned above, the Hf
596 isotopic compositions of bedloads and bank sediments are less radiogenic than those of
597 suspended loads sampled at the same location. Figure 9 shows clearly that this
598 difference increases with distance from river source. This is particularly clear for Ganges
599 sediments in which the Hf isotopic variation is less than 5 ϵ_{Hf} units near the border of the

600 Himalayas but reaches 12 ϵ_{Hf} units in the Bangladesh delta. Decoupling of Nd-Hf isotopes
601 exists at the front of the mountain range but becomes much more pronounced at the
602 river mouth. The increasing decoupling could result of enhanced differential chemical
603 weathering in the floodplain, with enhanced release of radiogenic Hf from high Lu-Hf
604 minerals in suspended loads. However, we do not observe any significant correlation
605 between ϵ_{Hf} values of suspended loads in the floodplain and the ratios used by Lupker et
606 al. (2012) to evaluate chemical weathering intensity (e.g. Na/Si, K/Si). Additionally, ϵ_{Hf}
607 values do not vary as a function of Lu/Hf or Ca/K from mountain front to delta, as would
608 be expected if radiogenic Hf were preferentially released by chemical weathering of high
609 Lu/Hf minerals (Bayon et al., 2006; Rickli et al., 2013). We therefore suggest that the
610 increasing decoupling of Nd and Hf isotopes along the Ganges floodplain is explained by
611 a change of river hydrodynamics, from a relatively turbulent regime that limited
612 mineralogical sorting within the water column at the mountain front, to a calmer regime
613 that allowed better mineral segregation in the floodplain and in the delta. Our
614 interpretation is further supported by the trace-element data shown in Figure 3. In
615 contrast to sediments from the Narayani, Karnali and Kosi Rivers (Figure 3f-g-h),
616 bedloads and bank sediments from the Bangladesh delta show significantly higher REE,
617 Th, U, Zr and Hf concentrations than suspended loads (Figure 3e). As shown in section
618 5.2.1, such differences reflect a higher degree of mineral sorting, in agreement with the
619 large differences of ϵ_{Hf} values observed between bedloads and suspended loads in the
620 Bangladesh delta (Figure 9).

621 The impact of mineralogical sorting processes on the overall chemistry of bed
622 and suspended loads from other large fluvial systems was highlighted in previous
623 studies (e.g. Gaillardet et al., 1995; Dupré et al., 1996; Bouchez et al., 2011). For example,
624 Bouchez et al. (2011) showed that mineral sorting of sediments explains a large part of

625 the chemical variability of bedloads and suspended loads in the Amazon River, another
626 large fluvial system in terms of sediment discharge into the world ocean (Milliman and
627 Meade, 1983; Milliman, 2001). Based on this observation, we suggest that Nd-Hf isotopic
628 decoupling of river sediments is not restricted to the Ganges but may occur in all detrital
629 sediments that are delivered into the ocean at the mouths of large rivers. Our results
630 may thus explain why detrital oceanic clays are displaced towards elevated ϵ_{Hf} relative
631 their ϵ_{Nd} values.

632

633 **5.4. Impact of continental fractionation on oceanic sediments**

634 Detrital sediments are transported from continent to ocean by rivers, glaciers or
635 winds. The present flux of riverine sediment from continent to ocean is estimated at
636 about $20 \times 10^{12} \text{ kg.y}^{-1}$ (Milliman and Meade, 1983; Hay, 1998 and references therein)
637 while the estimated present flux for glacial detritus ranges between 0.9 and 50×10^{12}
638 kg.y^{-1} and between 0.5 and $0.9 \times 10^{12} \text{ kg.y}^{-1}$ for dust (Hay, 1998 and references therein).

639 Here we evaluate whether the mineral sorting processes responsible for the Nd-
640 Hf isotopic decoupling in river sediments can explain the range of isotopic compositions
641 reported for worldwide terrigenous oceanic sediments (i.e. sand and clay). We do not
642 discuss the composition of biogenic sediments and Fe-Mn crusts and nodules because
643 their composition reflects that of dissolved Nd and Hf in seawater (e.g. Godfrey et al.,
644 2009; Rickli et al., 2009; Zimmerman et al., 2009; Stichel et al., 2012) and our study
645 focuses on particle loads. We use the vertical deviation from the Terrestrial Array ($\Delta\epsilon_{\text{Hf}}$)
646 (cf. Figure 8) to compare river and oceanic sediments. Part of the amplitude of $\Delta\epsilon_{\text{Hf}}$
647 values of worldwide sediments depends on the average age of the continental sources,
648 which can be evaluated using their ϵ_{Nd} values. Sediments eroded from old evolved crust
649 have very low ϵ_{Nd} values, and contain zircons with very low ϵ_{Hf} and phyllosilicates with

650 elevated ϵ_{Hf} . The range of $\Delta\epsilon_{\text{Hf}}$ should thus be very large. In contrast, erosion products of
651 younger crust have higher ϵ_{Nd} values, and the ϵ_{Hf} values of both zircons and
652 phyllosilicates are less extreme. The correction made to normalize the ϵ_{Hf} values of
653 sediments for the age effect is explained in the sketch shown in the inset of Figure 10.
654 We projected all initial Nd-Hf isotopic compositions of sediments to an arbitrary
655 intermediate ϵ_{Nd} value of -15 using straight lines that converge at an average depleted
656 mantle value of $\epsilon_{\text{Nd}} = +8.9$ and $\epsilon_{\text{Hf}} = +14.8$ (see caption of Figure 10). We then calculated
657 the vertical deviation ($\Delta\epsilon_{\text{Hf}}$) from the Terrestrial Array of all projected ϵ_{Hf} values to
658 obtain a parameter that is not influenced by the age effect. Such a projection allows us to
659 compare sediments from everywhere in the world. It can be noted that $\Delta\epsilon_{\text{Hf}}$ projected at
660 $\epsilon_{\text{Nd}} = -15$ is a robust parameter since the trend that we observe for river and oceanic
661 sediments does not significantly change when we choose other reasonable values for the
662 depleted mantle or for the ϵ_{Nd} value at which the projection is done.

663 The range of normalized $\Delta\epsilon_{\text{Hf}}$ is plotted in Figure 10 as a function of $\text{Al}_2\text{O}_3/\text{Zr}$
664 ratios for river and oceanic sediments. Like Vervoort et al. (2011), we selected $\text{Al}_2\text{O}_3/\text{Zr}$
665 instead of $\text{Al}_2\text{O}_3/\text{SiO}_2$ as proxy to the relative proportions of zircons vs. phyllosilicates
666 because the presence of biogenic silica in oceanic sediments affects $\text{Al}_2\text{O}_3/\text{SiO}_2$. Figure
667 10 shows that river and terrigenous oceanic sediments plot on the same trend in the
668 $\Delta\epsilon_{\text{Hf}}$ vs $\text{Al}_2\text{O}_3/\text{Zr}$ diagram. This suggests strongly that the decoupling of Nd and Hf
669 isotopic compositions in river and oceanic sediments results from the same process i.e.
670 mineral sorting, as suggested by Patchett et al. (1984). Oceanic sands and silts have $\Delta\epsilon_{\text{Hf}}$
671 values similar to those of the Himalayan river sediments analyzed in this study, while
672 oceanic clays have generally higher $\Delta\epsilon_{\text{Hf}}$ than the Himalayan suspended loads. This
673 shows that the decoupling observed in river sediments is amplified in the oceanic
674 environment. This arises because more than 90% of suspended matter transported by

675 rivers and nearly all bedloads are deposited in deltas and estuaries and never reach the
676 ocean (Lisitzin, 1996). Zircon is quantitatively removed from suspended matter during
677 transport from river mouth to open ocean (Stummeyer et al., 2002). Sorting processes at
678 the river/ocean interface thus amplify chemical fractionation observed in rivers: they
679 sequester zircon-rich sediments with unradiogenic Hf signature and deliver sediments
680 with radiogenic Hf to the ocean. Almost half of the Nd-Hf isotopic decoupling occurs in
681 rivers on continents (see Figure 10) but Hf-Nd fractionation most probably continues as
682 fine particles travel far from continents while coarser fractions sediment close to source
683 areas.

684 Our study on river sediments shows that mineral sorting controls Nd-Hf
685 decoupling in both continental and oceanic environments. The displacement of Nd-Hf
686 isotopic compositions of terrigenous oceanic clays above the Terrestrial Array in Nd-Hf
687 isotopic space is similar to that observed on continents during transport from sediment
688 sources to the open ocean. If the Nd-Hf isotopic compositions of oceanic sediments differ
689 from those of continental crust, their recycling in subduction zones should influence the
690 long-term evolution of the Earth's mantle. Indeed, Chauvel et al. (2008) showed that an
691 input of oceanic sediment is needed to explain both the Mantle Array and the mantle
692 sources of oceanic island basalts (OIB) and mid-ocean ridge basalts (MORB). Here, we go
693 one step further and propose that reinjection of oceanic sediments into the mantle has
694 shifted the Terrestrial Array towards higher ϵ_{Hf} at high ϵ_{Nd} values over Earth history and
695 has resulted in a systematic change of the slope of the array. To demonstrate this, we
696 estimate what would be the present-day Nd-Hf isotopic composition of the depleted
697 mantle if sediments and oceanic crust had not been recycled into the mantle through
698 Earth history, i.e., we calculate the composition of the depleted mantle without recycled
699 material. To do this, we need to know the present-day isotopic compositions of oceanic

700 sediments and oceanic crust that had formed at different times in the past (green and
701 pink lines in Figure 11, see Figure caption and Chauvel et al., 2008 for further details
702 about the calculations). As a rough guide, we also show the evolution of continental
703 crust and OIB through time (blue and orange lines in Figure 11). All parameters used to
704 calculate the evolution of these reservoirs are provided in Supplementary Table D. We
705 calculate an average composition for recycled oceanic crust and overlying sediments
706 (grey star in Figure 11) assuming a mixture of 10-20% oceanic sediment and 80-90%
707 oceanic crust (same procedure as in Chauvel et al., 2008) and an average age of 1.8 Ga
708 (i.e. the average age of the continental crust). Assuming that the present-day depleted
709 mantle – the source of MORB shown with a pink star in Figure 11 – consists in a mixture
710 of 90-95% depleted mantle without recycled material and 5-10% recycled material
711 (Chauvel et al., 2008; Sobolev et al., 2007), we can estimate the present-day isotopic
712 composition of the depleted mantle without recycled material. This is shown by the grey
713 field in Figure 11. The exact location of this field depends on the parameters used for
714 mixing calculations and in particular the Nd and Hf concentrations of the depleted
715 mantle. As we choose relatively high Nd and Hf concentrations (see Supplementary
716 Table D), our grey field defines the lowest possible range of ϵ_{Hf} and ϵ_{Nd} values. If we
717 choose lower Nd and Hf concentrations, for example those of Salters and Stracke (2004),
718 the depleted mantle without recycled material remains below the mantle array and has
719 a much more radiogenic isotopic composition, as shown by the dashed grey field in
720 Figure 11. The principal result is that the composition of the depleted mantle without
721 recycled material differs significantly from that of MORB and always lies at elevated ϵ_{Nd}
722 values below the Terrestrial array. This suggests that mantle-derived materials formed
723 in the earliest part of history probably had lower ϵ_{Hf} at any given ϵ_{Nd} than recently
724 formed material. Evaluating the impact of such a temporal change using the composition

725 of old continental crust and mantle is difficult because data on old and well-constrained
726 materials are not numerous. Additional Nd and Hf isotopic data on carefully selected
727 rocks with ages covering the Earth history could provide further constraints.

728 The Terrestrial Array that describes the overall relationship between Nd and Hf
729 isotopes is based on all available terrestrial data (Vervoort et al., 1999; Vervoort et al.,
730 2011). Not only does it represent a mixture of materials formed at different times from
731 the mantle but it also is strongly influenced by the large number of analyses obtained on
732 recent basalts. We thus suggest that the slightly positive ϵ_{Hf} intercept and, more
733 significantly, the large envelope of data surrounding the best-fit line through the
734 Terrestrial Array could be due to the temporal increase of incongruence of the two
735 isotopic systems.

736

737 **6. Summary and Conclusions**

738 This paper describes the first comprehensive study of Nd and Hf isotopic
739 compositions in bedloads, suspended loads and bank sediments sampled along one of
740 the largest rivers, the Ganges. Because source heterogeneity does not significantly affect
741 the isotopic composition of the studied sediments, we could demonstrate that mineral
742 sorting processes control the chemistry of Ganges river sediments and account for their
743 entire Hf isotopic variability ($>10 \epsilon_{\text{Hf}}$). Bedload and bank sediments always have lower
744 ϵ_{Hf} than suspended loads sampled at the same location. As suggested by Patchett et al.
745 (1984) for detrital oceanic sediments, coarse-grained sediments fall next to the
746 Terrestrial Array in Hf-Nd isotopic space while fine-grained sediments display
747 increasing decoupling of the two isotopic systems as sediment grain size decreases. We
748 attribute these features to hydraulic sorting of minerals during transport on the
749 floodplain, with the selective deposition of zircons that host significant amounts of

750 unradiogenic Hf. Zircons dominate the Hf isotopic budget of coarse sediments while
751 their contribution to the Hf isotopic budget of finer sediments is balanced by minerals
752 with more radiogenic Hf such as phyllosilicates.

753 We show that the Nd-Hf isotopic decoupling increases from mountain source to
754 the estuary where it reaches its maximum. Decoupling of Nd and Hf isotopes in river
755 sediments and oceanic clays is caused by the same processes, i.e., mineral sorting during
756 transport. We speculate that transport of fine particles over long distances in the open
757 ocean amplifies the signal. Finally, the recycling of oceanic sediments with anomalous
758 Nd-Hf isotopic compositions into the mantle, would have slowly shifted the overall
759 isotopic composition of the Earth's mantle towards more radiogenic Hf values.

760

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769

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1023

1024 **Table Captions**

1025 **Table 1:** Nf and Hf isotopes in river sediments

1026 *Footnotes:* Re-run samples correspond to run duplicates and Dup samples stand for
1027 complete analytical duplicates. Uncertainties ($\pm 2\sigma$) indicated in the Table are in-run
1028 errors. ϵ_{Nd} and ϵ_{Hf} values were calculated using the CHUR composition published by
1029 (Bouvier et al., 2008).

1030

1031 **Figure Captions**

1032 **Figure 1:** Map of the Ganges fluvial system showing the main sampling sites for the river
1033 sediments analyzed in this study.

1034 *Footnotes:* Blue stars indicate the locations of samples from the Ganges River, red stars
1035 those from the Karnali River, purple stars those from the Narayani river, yellow stars
1036 those from the Kosi River. White, light and dark grey stars indicate the locations of the
1037 samples from Nepalese rivers draining the Lesser Himalaya units, the High Himalayan

1038 Crystalline units and the Tethyan Sedimentary Series, respectively. The precise locations
1039 can be found in Pierson-Wickmann et al. (2000) and Garzanti et al. (2007) while their
1040 GPS coordinates are provided in Supplementary Table B. Detailed locations of the 10
1041 samples recovered in the Marsyandi River (pink star) can be found in Garzanti et al.
1042 (2007) and GPS coordinates are provided in Supplementary Table B.

1043

1044 **Figure 2: a)** Variability of SiO_2 and Al_2O_3 contents in river sediments. **b)** Histogram
1045 showing the frequency of $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios for the studied bedloads, banks and
1046 suspended loads.

1047 *Footnotes:* Red and green stars correspond to the average composition of the Ganges
1048 bedload and surface suspended load in Bangladesh, as published by Lupker et al. (2011).
1049 The composition of the average Upper Continental Crust (Rudnick and Gao, 2003) is
1050 shown as indication.

1051

1052 **Figure 3:** Trace element patterns normalized to the average composition of the upper
1053 continental crust (UCC) of Rudnick and Gao (2003) for sediments sampled in rivers
1054 draining single geological units (a-b-c) as well as in the Marsyandi (d), the Ganges (e),
1055 the Narayani (f), the Karnali (g) and the Kosi rivers (h).

1056

1057 **Figure 4:** ϵ_{Hf} versus ϵ_{Nd} diagram showing river sediments sampled in the Himalayan
1058 Range i.e. in the Marsyandi River and in small Nepalese rivers draining only the Tethyan
1059 Sedimentary Series, the High Himalayan Crystalline and the Lesser Himalaya.

1060 *Footnotes:* Nd-Hf isotopic compositions of worldwide oceanic sediments are also shown.

1061 Data for Fe-Mn crusts and nodules are from Albarède et al. (1997); Godfrey et al. (1997);

1062 Albarède et al. (1998); David et al. (2001); van De Flierdt et al. (2006); oceanic clays and

1063 biogenic muds are from White et al. (1986); Ben Othman et al. (1989); McLennan et al.
1064 (1990); Pearce et al. (1999); Vervoort et al. (1999); Woodhead et al. (2001); Vlastélic
1065 (2005); Prytulak et al. (2006); Carpentier et al. (2008); Bayon et al. (2009); Carpentier
1066 et al. (2009); Chauvel et al. (2009); Vervoort et al. (2011) and oceanic silts and sands are
1067 from McLennan et al. (1990); Vervoort et al. (1999); Prytulak et al. (2006); Carpentier et
1068 al. (2008); Bayon et al. (2009); Carpentier et al. (2009); Vervoort et al. (2011). The
1069 present-day Terrestrial Array is that of Vervoort et al. (2011). The histogram along the
1070 ϵ_{Nd} axis shows ϵ_{Nd} values reported for Himalayan bedrocks from the three main
1071 geological units (Parrish and Hodges, 1996; Ahmad et al., 2000; Robinson et al., 2001;
1072 Richards et al., 2005). All ϵ_{Nd} and ϵ_{Hf} values were recalculated using the CHUR
1073 composition published by Bouvier et al. (2008).

1074

1075 **Figure 5:** ϵ_{Hf} versus ϵ_{Nd} diagram showing data obtained on sediments sampled in the
1076 floodplain i.e. in the Ganges and its main tributaries.

1077 *Footnotes:* Same sources of data as in Figure 4. ϵ_{Nd} and ϵ_{Hf} values calculated using the
1078 CHUR composition published by Bouvier et al. (2008).

1079

1080 **Figure 6:** Downstream evolution of ϵ_{Nd} and ϵ_{Hf} in sediments along the Marsyandi River
1081 located in the Himalayan Range.

1082 *Footnotes:* The river profile and the main geological units on which the river flows are
1083 shown together with the location of two towns, Tal and Besi Shahar.

1084

1085 **Figure 7:** Variations of ϵ_{Hf} and ϵ_{Nd} as a function of Al_2O_3/SiO_2 ratio used as a grain-size
1086 proxy for the Ganges, the Narayani, the Karnali and the Kosi river sediments.

1087 *Footnotes:* The color lines link bedloads and banks to their corresponding suspended
1088 loads sampled at the same location at different depths in the water column.

1089

1090 **Figure 8:** $\Delta\epsilon_{\text{HF}}$ vs. $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio used as grain-size proxy for the Ganges, the Narayani,
1091 the Karnali and the Kosi river sediments.

1092 *Footnotes:* The sketch at the bottom right corner shows how vertical deviations ($\Delta\epsilon_{\text{HF}}$)
1093 from the present-day Terrestrial Array of Vervoort et al. (2011) are calculated.

1094

1095 **Figure 9:** Downstream changes of ϵ_{Nd} and ϵ_{HF} in sediments from the Ganges and its
1096 major tributaries in the floodplain, from the Himalayan Mountain front to the Bengal
1097 delta.

1098 *Footnotes:* Same symbols and colors as in previous figures. Color lines link bedloads and
1099 banks to their corresponding suspended loads sampled at the same location at different
1100 depths in the water column. The topography of the river profile is shown in brown.

1101

1102 **Figure 10:** $\Delta\epsilon_{\text{HF}}$ projected at $\epsilon_{\text{Nd}}=-15$ plotted as a function of $\text{Al}_2\text{O}_3/\text{Zr}$ ratios for river
1103 sediments analyzed in this study and worldwide oceanic terrigenous sediments (oceanic
1104 sands, silts and clays).

1105 *Footnotes:* The sketch shown in the inset explains how all measured and published ϵ_{HF}
1106 are projected to $\epsilon_{\text{Nd}}=-15$ to calculate the vertical deviation from the present-day
1107 Terrestrial Array of Vervoort et al. (2011): $\Delta\epsilon_{\text{HF}}$ that is plotted in the main panel. The ϵ_{Nd}
1108 value of the depleted mantle pole was compiled using data from the PETDB database
1109 and its ϵ_{HF} was calculated by projecting this ϵ_{Nd} value on the Terrestrial Array of
1110 Vervoort et al. (2011). In the main panel, colors and symbols for river sediments
1111 analyzed in this study are the same as in previous figures. Data for oceanic sands, silts

1112 and clays correspond to terrigenous sediments poor in volcanic ashes and are from
1113 Plank et al. (2007); Carpentier et al. (2008); Carpentier et al. (2009); Chauvel et al.
1114 (2009); Vervoort et al. (2011). ϵ_{Hf} and ϵ_{Nd} of terrigenous sediments have been corrected
1115 for subsequent decay after their deposition age.

1116

1117 **Figure 11:** Present-day Nd-Hf isotopic compositions of the different Earth reservoirs
1118 and our estimated values for the depleted mantle without recycled material.

1119 *Footnotes:* The present-day Nd-Hf isotopic compositions of oceanic sediments are from
1120 Chauvel et al. (2008). Present-day Nd isotopic compositions of MORB and OIB are from
1121 Gale et al. (2013) and PetDB database, respectively. Hf isotopic compositions of MORB
1122 and OIB were calculated from their Nd isotopic composition to lie on the present-day
1123 Terrestrial Array of Vervoort et al. (2011). Colored lines show the present-day isotopic
1124 compositions of oceanic sediments, continental crust, OIB and MORB that had formed at
1125 different times in the past. To model these evolutions, we assumed a linear relationship
1126 between the present-day isotopic compositions of the reservoirs and the Nd-Hf isotopic
1127 ratios of the Earth at 4.55 Ga. Oceanic sediments, continental crust, OIB and MORB are
1128 then modeled to evolve through time following their own Sm/Nd and Lu/Hf ratios (see
1129 Supplementary Table D). The grey star shows the average Nd-Hf isotopic composition of
1130 the material being recycled in the mantle. It is calculated using a binary mixing between
1131 10-20% of 1.8 Ga oceanic sediment and 80-90% of 1.8 Ga oceanic crust (black mixing
1132 line with ticks indicating the proportion of sediment in the mixture). The field of
1133 present-day values for the depleted mantle without recycled material (grey field) was
1134 calculated assuming that the present-day depleted mantle (average MORB) results of a
1135 mixture of 5-10% recycled material and 90-95% depleted mantle without recycled
1136 material (grey mixing line). The dashed grey field shows the composition of the same

1137 reservoir if the MORB source has very low Nd and Hf concentrations (i.e. 0.713 ppm for
1138 Nd and 0.199 ppm for Hf; Salters and Stracke, 2004).

Table 1: Hf and Nd isotopes in sediments*From rivers draining monolithologic basins in the Himalayan range*

Sample Name	Locality	River	Type of sediment	$^{176}\text{Hf}/^{177}\text{Hf} \pm 2\sigma$	ϵHf	$^{143}\text{Nd}/^{144}\text{Nd} \pm 2\sigma$	ϵNd
Lesser Himalaya units							
R 94-12	Sarmi, Nepal	Tributary of the Bheri River	Bedload	0.281956	11 -29.3	0.511422	11 -23.6
MO 102	Darondi/Marsel, Nepal	Marsel Khola	Bank	0.281798	5 -34.9	0.511436	9 -23.3
MO 112	Bhuri Gandaki/Isul, Nepal	Isul Khola	Bank	0.281646	5 -40.3	0.511380	6 -24.4
MO 112 re-run						0.511379	6 -24.4
PB 33	Nepal	Ghara khola	Suspended Load	0.281972	9 -28.8	0.511577	8 -20.5
PB 37	Nepal	Beg Khola	Bank	0.281659	13 -39.8	0.511488	5 -22.3
PB 37 re-run						0.511488	5 -22.3
MO 207	Waling, Nepal	Andi Khola	Bedload	0.281672	20 -39.4	0.511343	7 -25.1
MO 207 re-run				0.281663	7 -39.7		
High Himalayan Crystalline units							
MO 73	Nepal	Tributary of the Chepe khola	Bank	0.282243	12 -19.2	0.511724	10 -17.7
MO 50	Nepal	Chepe khola	Bank	0.282427	13 -12.7	0.511968	5 -12.9
MO 59	Nepal	Chepe khola	Bank	0.282374	9 -14.5	0.511953	9 -13.2
MO 59 re-run				0.282348	5 -15.4		
KN 83	Ghyangphedi, Nepal	Tadi khola	Bedload	0.282156	10 -22.2	0.511811	6 -16.0
Tethyan Sedimentary Series							
MO 504	Tukche, Nepal	Yamkim khola	Suspended Load	0.282235	8 -19.5	0.511730	6 -17.6
NAG 22	Pedi, Nepal	Marsyandi (10 km away from the River Source)	Bedload	0.282175	9 -21.6	0.511779	5 -16.6
NAG 22 re-run					0	0.511778	6 -16.6
MAR-50	Temang, Nepal	Marsyandi (60 km away from the River Source)	Bedload	0.282138	8 -22.9	0.511760	6 -17.0
MAR-50 re-run				0.282146	11 -22.6	0.511756	5 -17.0
MAR-40	Sabche, Nepal	Sabche khola	Bedload	0.282114	8 -23.7	0.511693	6 -18.3
MAR-40 Dup						0.511695	8 -18.2

From the Marsyandi River in the Himalayan range

Sample Name	Locality	Distance from River Source (km)	Type of sediment	$^{176}\text{Hf}/^{177}\text{Hf} \pm 2\sigma$	ϵHf	$^{143}\text{Nd}/^{144}\text{Nd} \pm 2\sigma$	ϵNd
MAR-55	Tal, Nepal	72	Bedload	0.282202	9 -20.6	0.511798	7 -16.2
MAR-58	Chamje, Nepal	75	Bedload	0.282151	8 -22.4	0.511803	7 -16.1
MAR-70	Besi Sahar, Nepal	104	Bedload	0.282178	8 -21.5	0.511813	8 -15.9
MAR-31	Philesangu, Nepal	111	Bedload	0.282065	8 -25.5	0.511816	6 -15.9
MAR-29	Suribar, Nepal	119	Bedload	0.282207	8 -20.4	0.511715	6 -17.8
MAR-29 re-run						0.511724	8 -17.7
MAR-19	Turture, Nepal	133	Bedload	0.282222	6 -19.9	0.511841	6 -15.4
MAR-19 re-run				0.282218	9 -20.1		
MAR-12	Anpu, Nepal	161	Bedload	0.282126	9 -23.3	0.511803	5 -16.1
MAR-10	Lobrang, Nepal	166	Bedload	0.282189	6 -21.1	0.511724	7 -17.7
MAR-10 Dup	Lobrang, Nepal	166	Bedload	0.282219	11 -20.0	0.511709	7 -18.0

From the Ganga river and its major tributaries in the floodplain

Sample Name	Locality	Distance from River Source (km)	Type of sediment	$^{176}\text{Hf}/^{177}\text{Hf} \pm 2\sigma$	ϵHf	$^{143}\text{Nd}/^{144}\text{Nd} \pm 2\sigma$	ϵNd
Ganga River							
BR 931	Devrapayag, India	260	Suspended Load	0.282118	7 -23.6	0.511818	8 -15.8
BR 932	Devrapayag, India	260	Suspended Load	0.282070	6 -25.3	0.511724	5 -17.7
BR 922	Rishikesh, India	260	Suspended Load	0.282180	6 -21.4	0.511706	6 -18.0
BR 922 re-run						0.511715	7 -17.9

Sample Name	Locality	Distance from River Source (km)	Type of sediment	$^{176}\text{Hf}/^{177}\text{Hf} \pm 2\sigma$	ϵHf	$^{143}\text{Nd}/^{144}\text{Nd} \pm 2\sigma$	ϵNd		
BR 924	Rishikesh, India	260	Bank	0.282053	7	-25.9	0.511737	7	-17.4
BR 943	Kanpur, India	800	Suspended Load	0.282217	8	-20.1	0.511829	6	-15.6
BR 946	Kanpur, India	800	Bedload	0.282055	5	-25.8	0.511780	8	-16.6
BR 946 Dup	Kanpur, India	800	Bedload	0.282004	8	-27.6	0.511728	5	-17.6
BGP 6	Rajshahi, India	2570	Bedload	0.282027	11	-26.8	0.511758	9	-17.0
BGP 6 re-run		2570					0.511752	6	-17.1
BGP 6 Dup	Rajshahi, India	2570	Bedload	0.282009	7	-27.5	0.511749	7	-17.2
BR 415	Harding bridge, Bangladesh	2640	Suspended Load	0.282241	9	-19.2	0.511752	6	-17.1
BR 414	Harding bridge, Bangladesh	2640	Suspended Load	0.282226	6	-19.8	0.511736	8	-17.4
BR 413	Harding bridge, Bangladesh	2640	Suspended Load	0.282205	6	-20.5	0.511748	6	-17.2
BR 413 re-run				0.282198	5	-20.8			
BR 412	Harding bridge, Bangladesh	2640	Suspended Load	0.282209	6	-20.4	0.511745	7	-17.3
BR 411	Harding bridge, Bangladesh	2640	Suspended Load	0.282133	7	-23.0	0.511755	6	-17.1
BR 411 Dup	Harding bridge, Bangladesh	2640	Suspended Load				0.511723	7	-17.7
BR 418	Harding bridge, Bangladesh	2640	Bedload	0.281992	11	-28.1	0.511719	5	-17.8
BR 8252	Harding bridge, Bangladesh	2640	Bedload				0.511788	11	-16.4
BR 8253	Harding bridge, Bangladesh	2640	Suspended Load	0.282268	7	-18.3	0.511767	8	-16.8
BR 717	Harding bridge, Bangladesh	2640	Bedload	0.281930	6	-30.2	0.511769	7	-16.8
BR 717 re-run				0.281944	8	-29.8			
Karnali River									
PB 79	Chisapani, Nepal	260	Suspended Load	0.282155	8	-22.3	0.511721	6	-17.7
PB 79 re-run							0.511717	6	-17.8
PB 80	Chisapani, Nepal	260	Bank	0.282033	8	-26.6	0.511775	8	-16.7
BR 8113	Revelganj, India	850	Suspended Load	0.282206	6	-20.5	0.511758	7	-17.0
BR 8113 re-run							0.511744	6	-17.3
BR 8113 Dup	Revelganj, India	850	Suspended Load	0.282192	6	-21.0			
BR 8115	Revelganj, India	850	Bedload	0.282027	8	-26.8	0.511684	6	-18.5
BR 8115 Dup	Revelganj, India	850	Bedload				0.511714	9	-17.9
BR 8115 Ter	Revelganj, India	850	Bedload				0.511712	5	-17.9
Kosi River									
LO 762	Chatara, Nepal	180	Suspended Load	0.282155	6	-22.3	0.511714	5	-17.9
LO 762 re-run				0.282162	9	-22.0			
LO 763	Chatara, Nepal	180	Bank	0.282049	5	-26.0	0.511689	7	-18.4
LO 763 Dup	Chatara, Nepal	180	Bank	0.282040	7	-26.3			
PB 69	Chatara, Nepal	440	Suspended Load	0.282123	5	-23.4	0.511601	8	-20.1
PB 70	Chatara, Nepal	440	Bank	0.282064	9	-25.5	0.511649	8	-19.1
BR 331	Dumarighat, India	440	Bank	0.282136	7	-23.0	0.511656	7	-19.0
BR 332	Dumarighat, India	440	Bank	0.282125	6	-23.3	0.511722	9	-17.7
BR 332 re-run				0.282122	6	-23.5			
Narayani River									
LO 757	Narayanghat, Nepal	200	Suspended Load	0.282112	7	-23.8	0.511711	7	-17.9
LO 756	Narayanghat, Nepal	200	Suspended Load	0.282114	6	-23.7	0.511686	6	-18.4
LO 754	Narayanghat, Nepal	200	Suspended Load	0.282085	5	-24.7	0.511681	6	-18.5
LO 755	Narayanghat, Nepal	200	Suspended Load	0.282092	10	-24.5	0.511662	7	-18.9
LO 755 Dup	Narayanghat, Nepal	200	Suspended Load	0.282067	10	-25.4			
LO 758 C	Narayanghat, Nepal	200	Suspended Load	0.282135	4	-23.0	0.511661	5	-18.9
PB 54	Narayanghat, Nepal	200	Suspended Load	0.282055	7	-25.8	0.511661	5	-18.9
LO 1002	Narayanghat, Nepal	200	Suspended Load	0.282129	15	-23.2	0.511712	8	-17.9
LO 1002 re-run				0.282125	12	-23.3	0.511743	11	-17.3
LO 1007	Narayanghat, Nepal	200	Suspended Load	0.282007	12	-27.5	0.511655	13	-19.0
LO 1007 Dup	Narayanghat, Nepal	200	Suspended Load	0.282001	6	-27.7	0.511632	11	-19.5
BR 8106	Hajipur, India	550	Suspended Load	0.282149	6	-22.5	0.511633	6	-19.4
BR 8107	Hajipur, India	550	Bedload	0.282100	5	-24.2	0.511734	7	-17.5

Figure 1

Figure 1

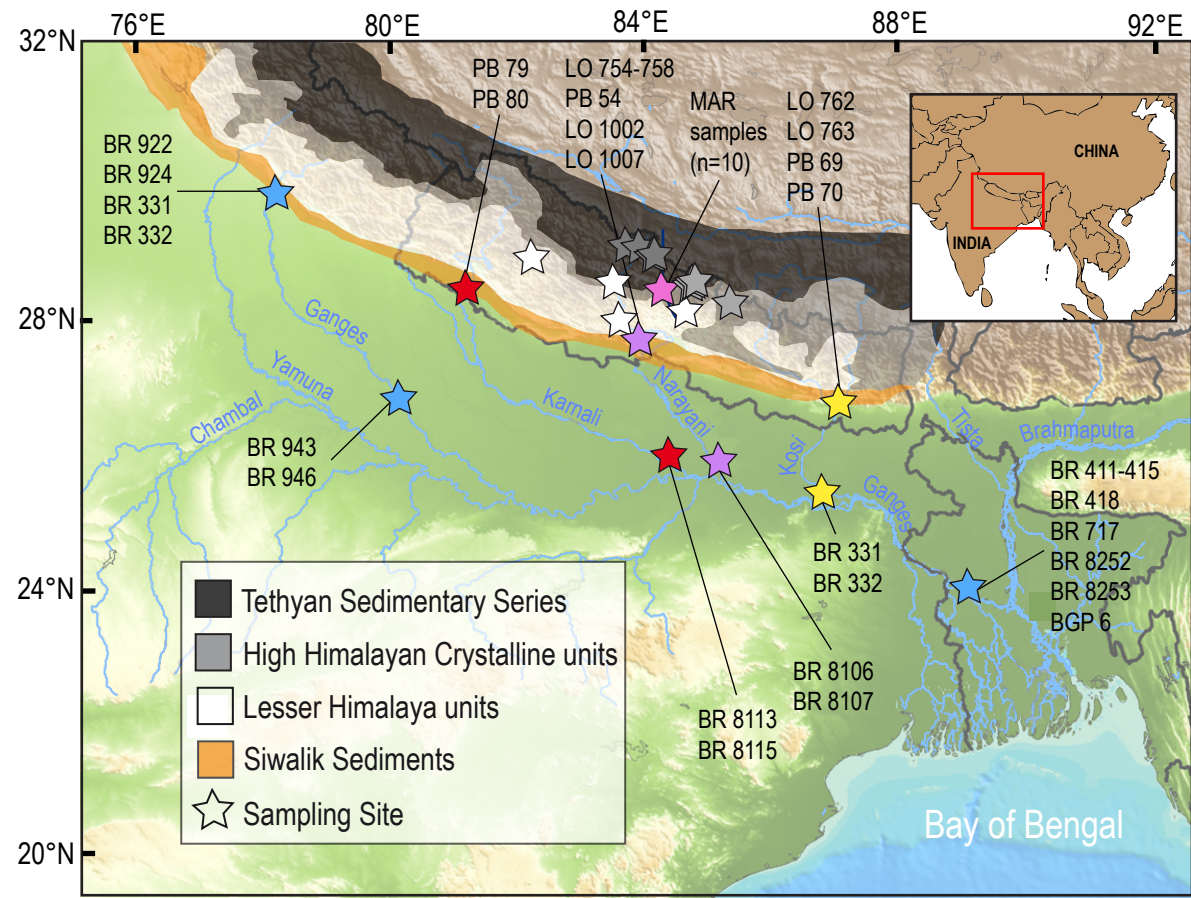


Figure 2
Figure 2

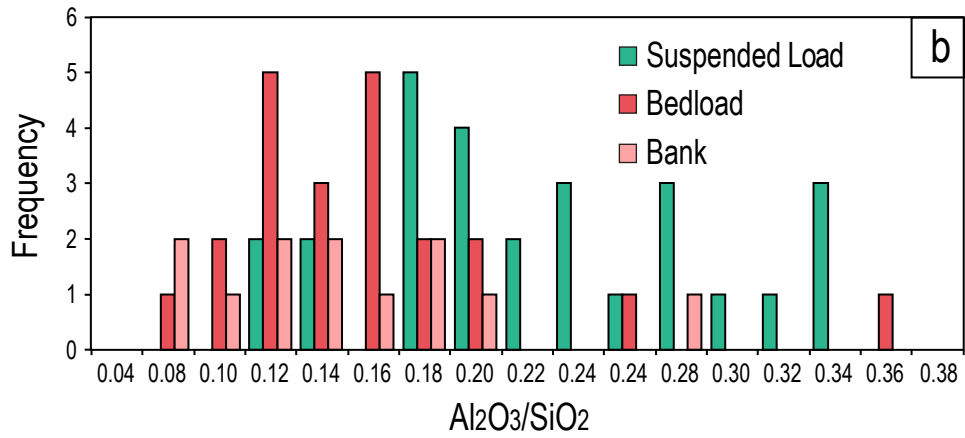
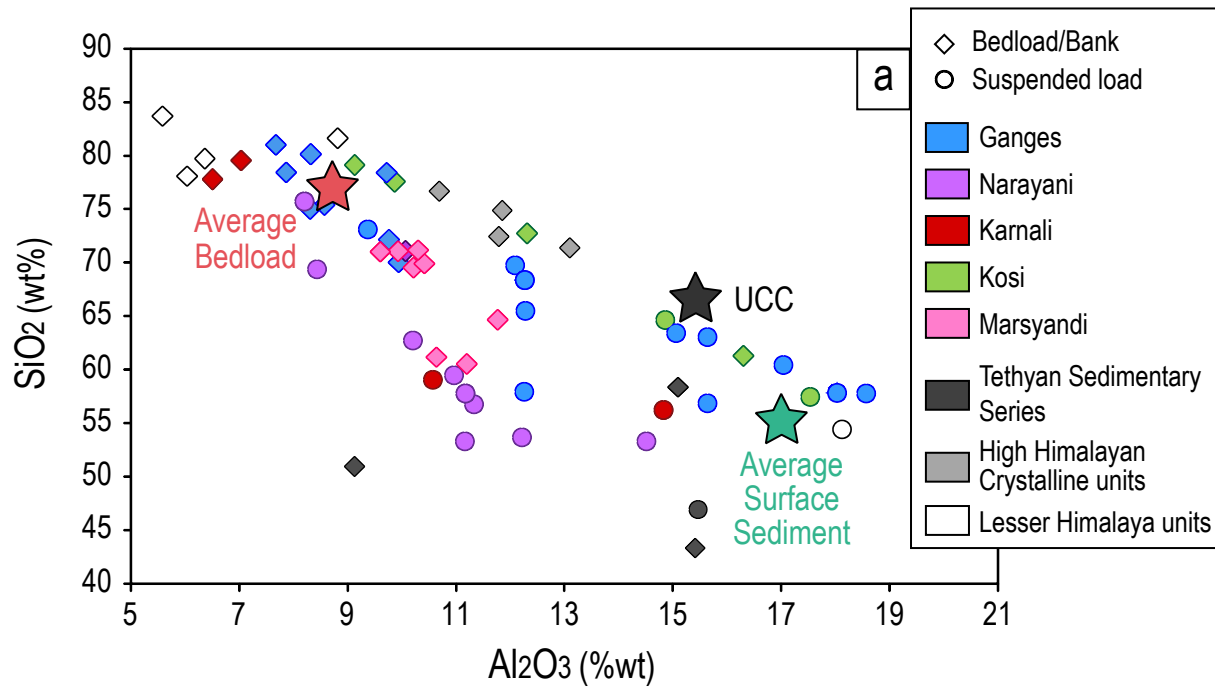


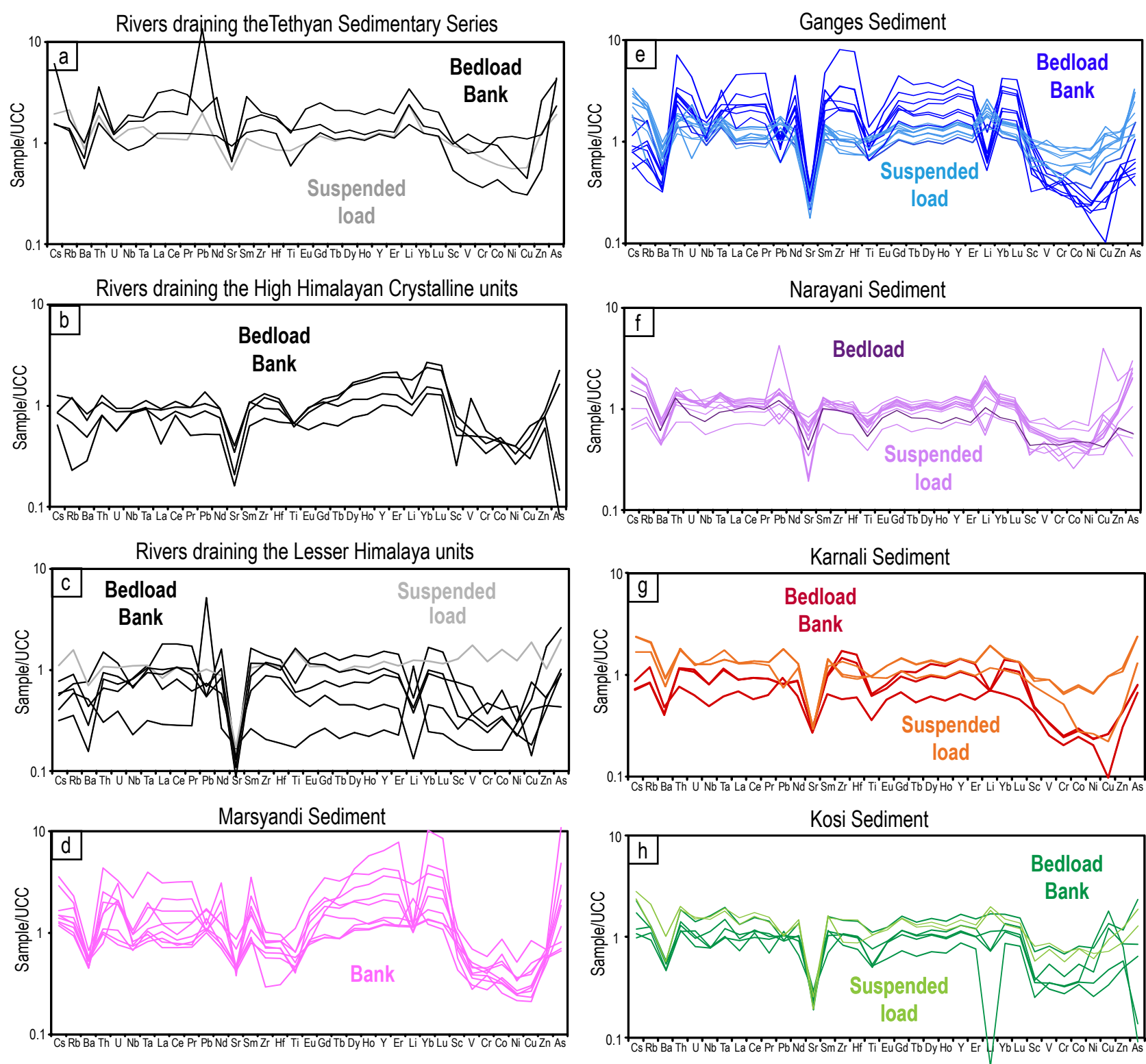
Figure 3

Figure 4

Figure 4

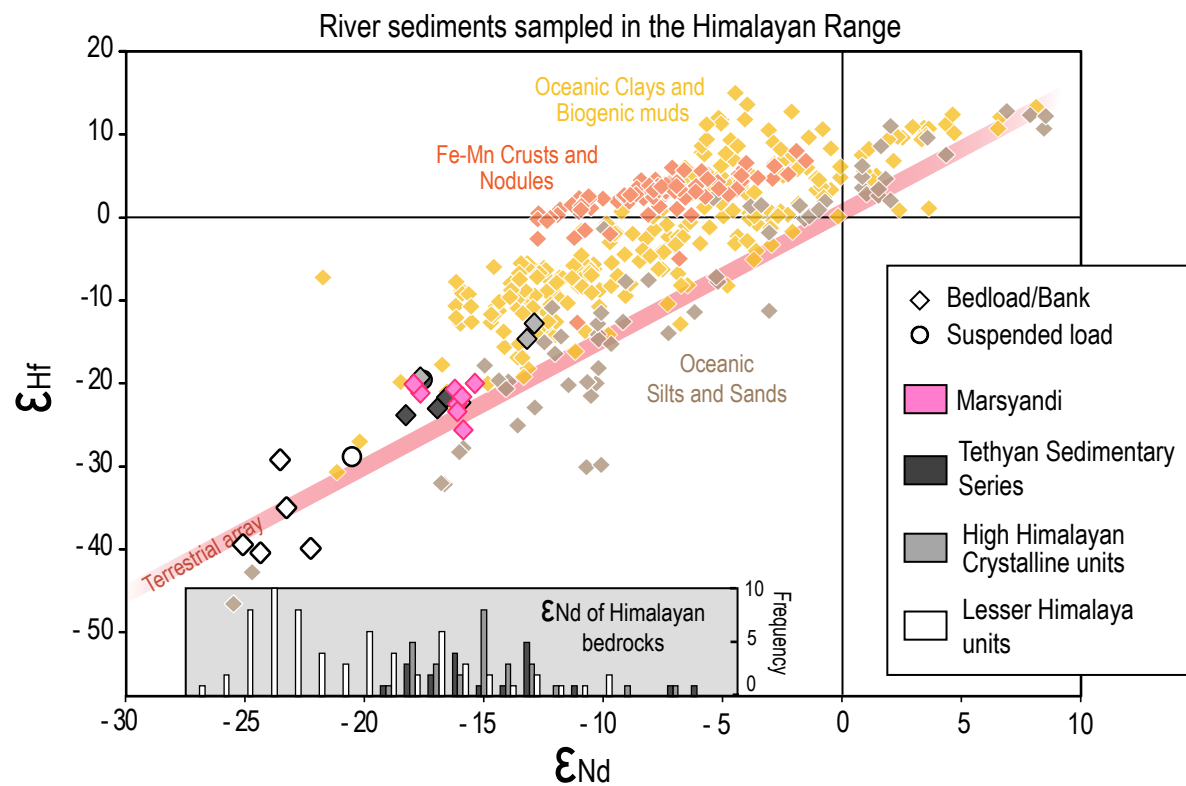


Figure 5
Figure 5

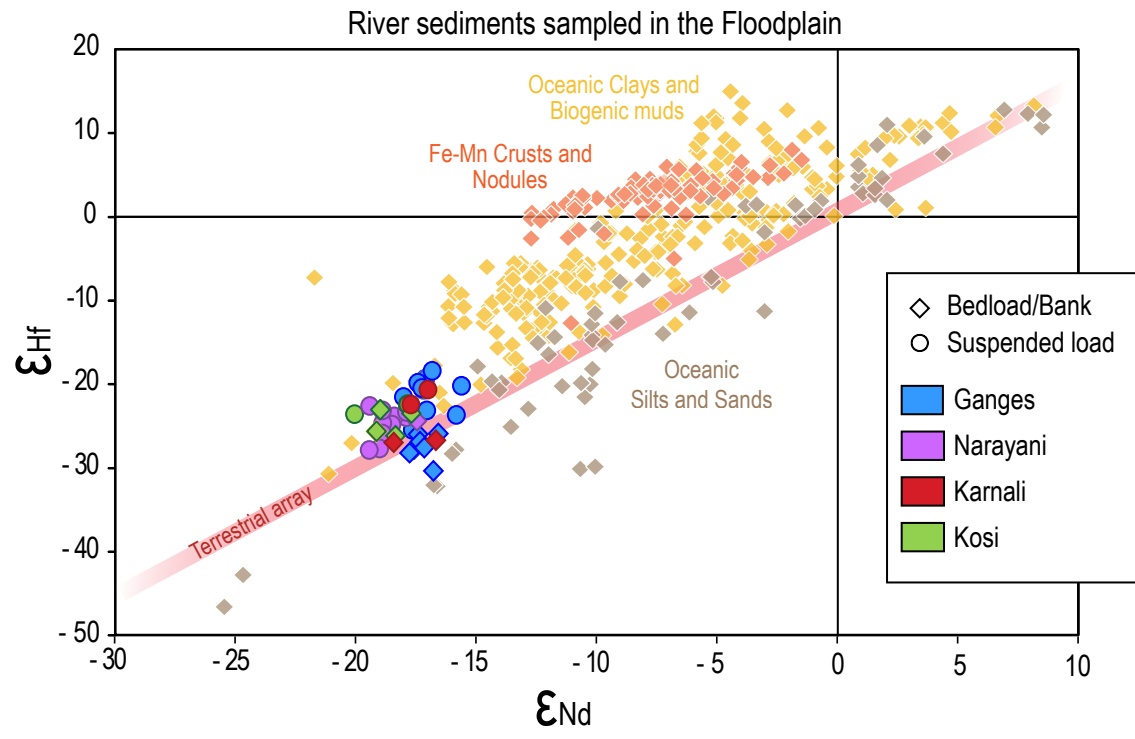


Figure 6

Figure 6

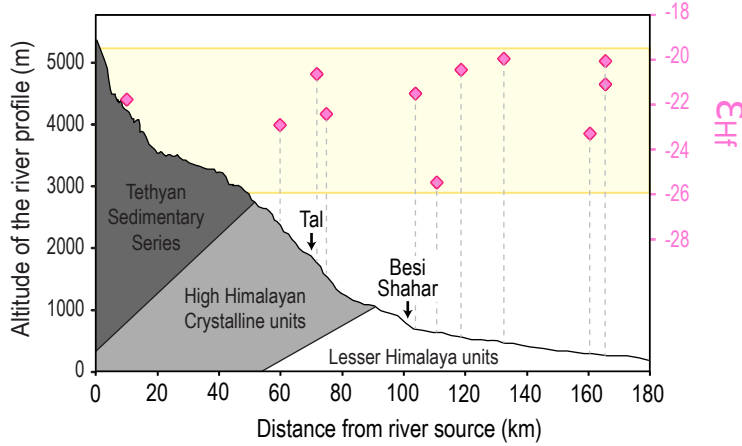
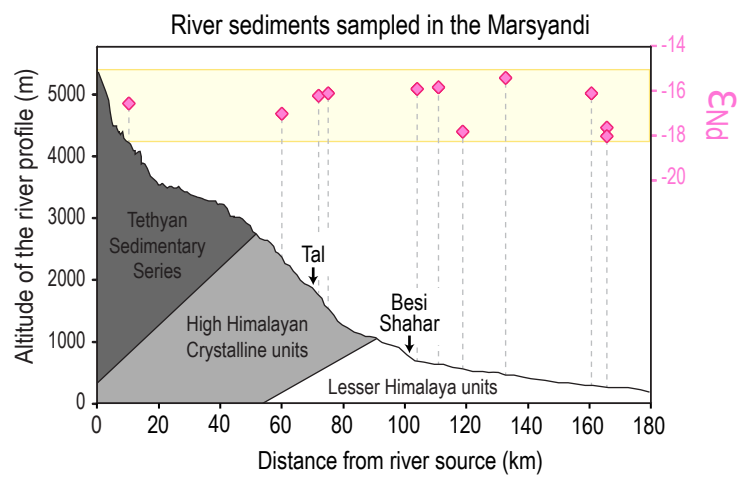


Figure 7

Figure 7

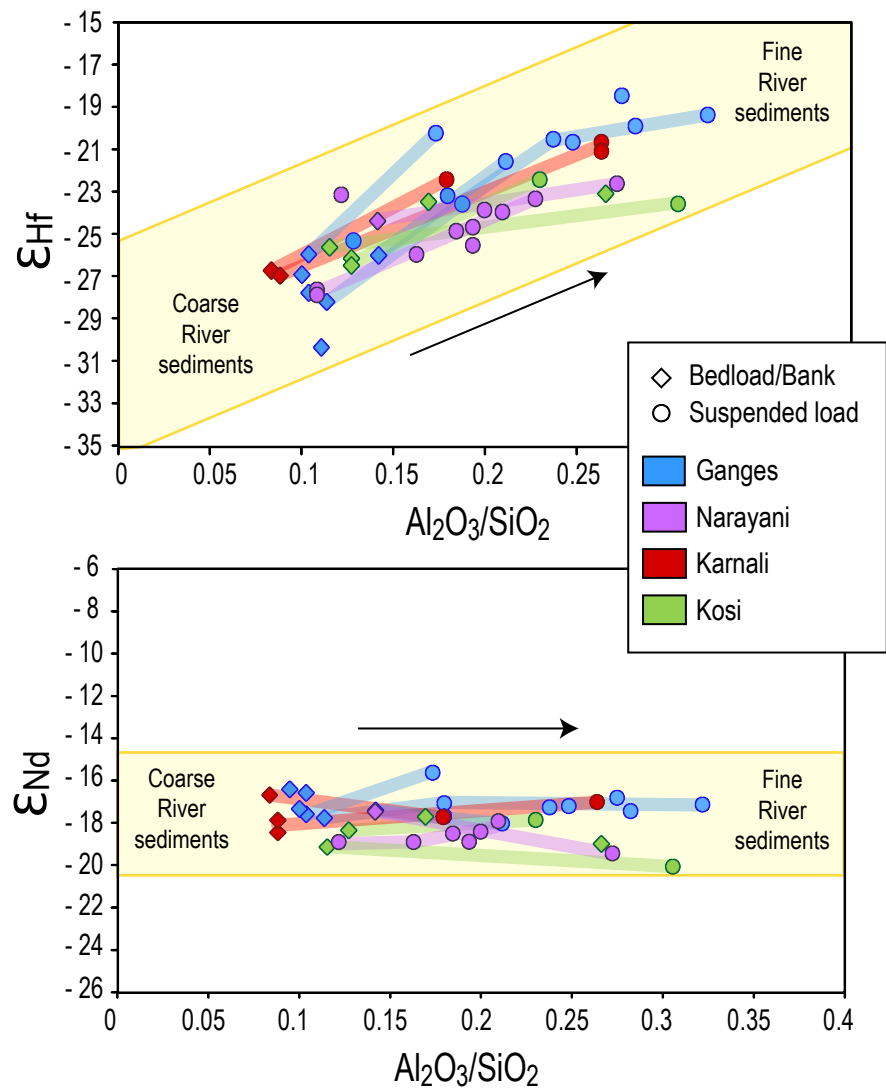


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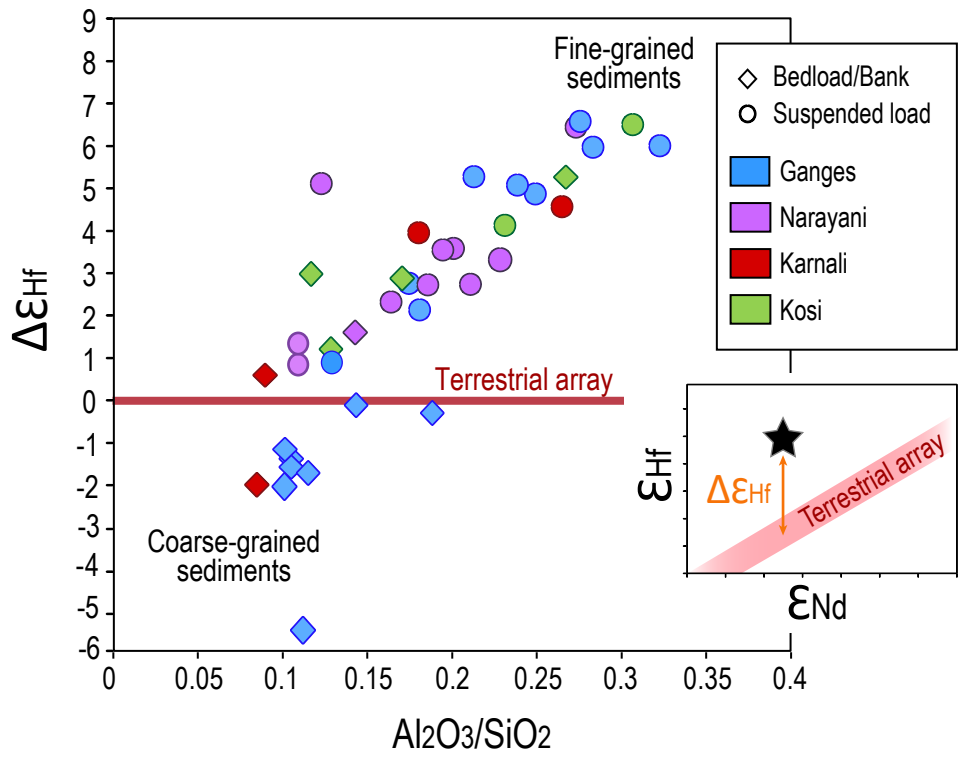


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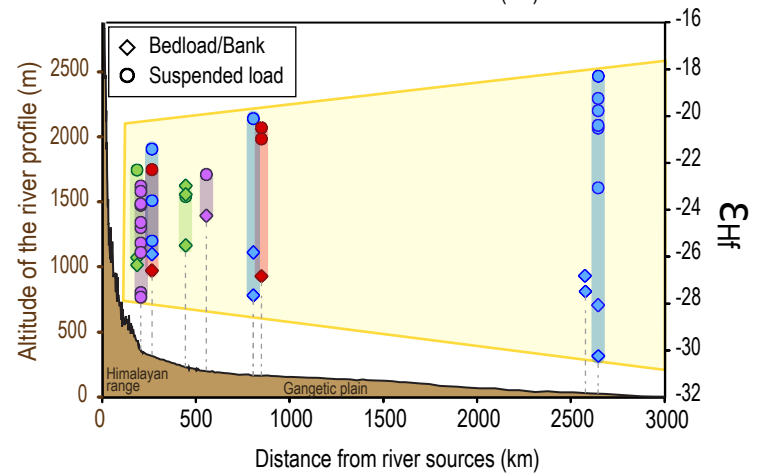
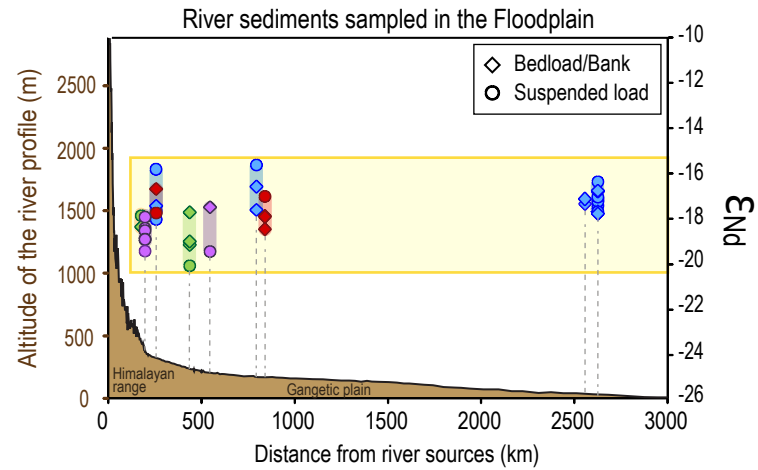


Figure 10

Figure 10

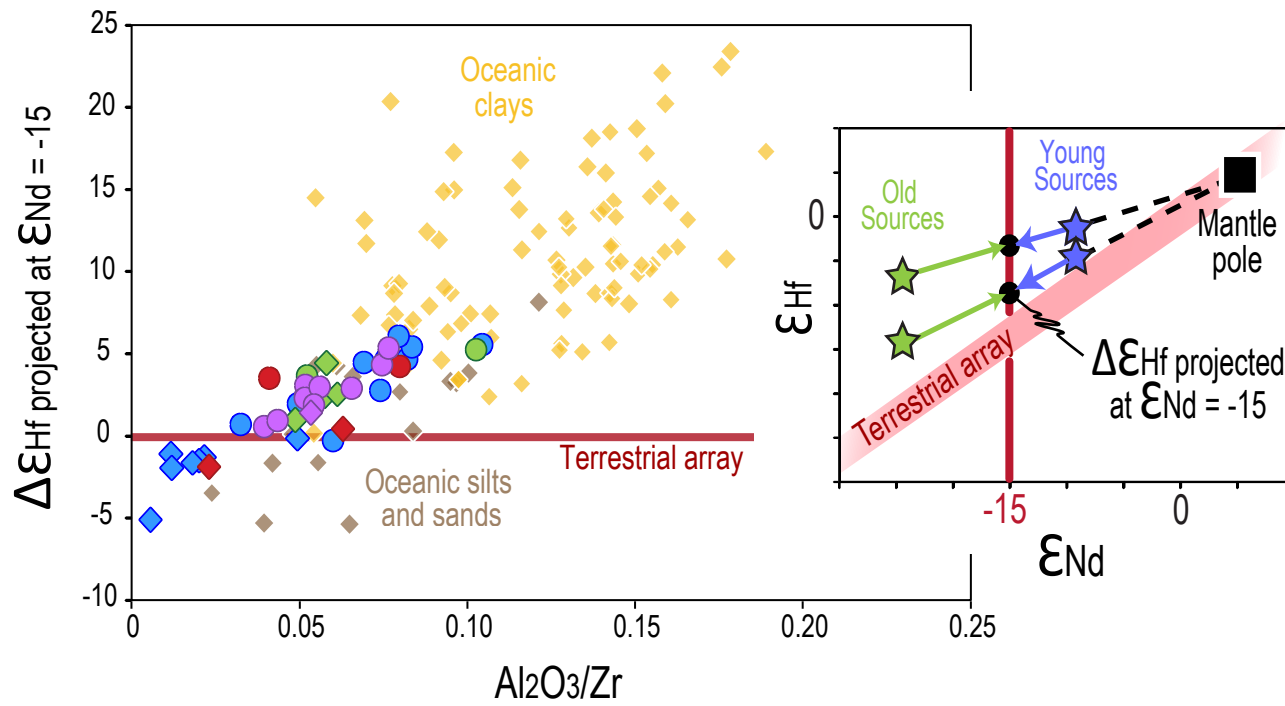
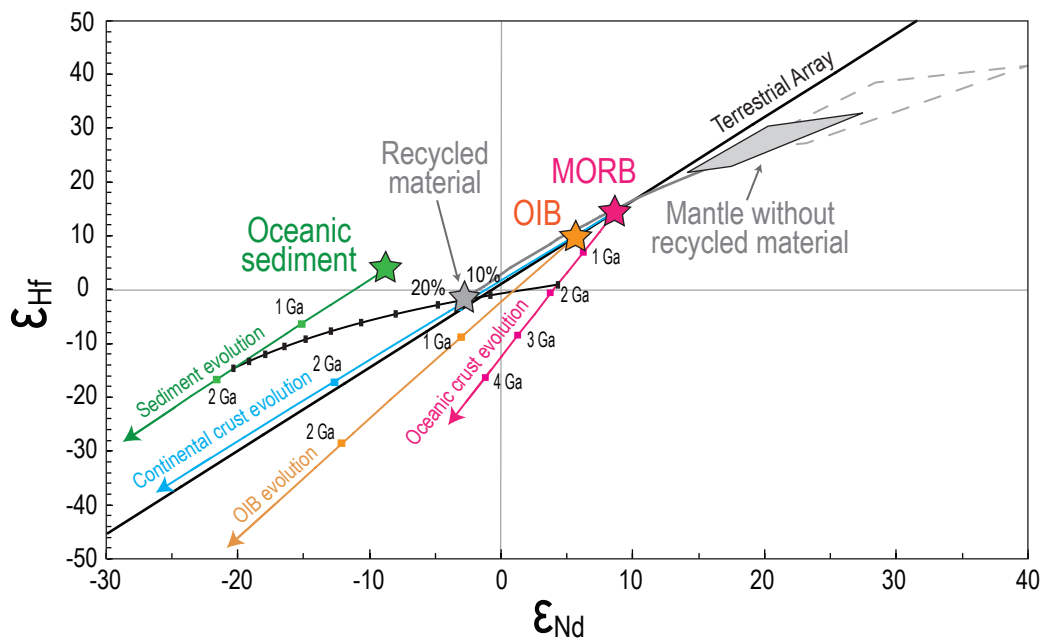


Figure 11



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

Surface suspended loads (<3m)			Deep suspended loads (>8m)			Bedloads		
	Average	2 σ		Average	2 σ		Average	2 σ
Quartz	34.3	22.8	Quartz	48.7	10.6	Quartz	55.4	14.9
K-feldspar	2.6	1.6	K-feldspar	5.7	2.3	K-Feldspar	6.6	3.1
Albite	4.6	2.4	Albite	5.9	4.0	Plagioclase	6.0	2.9
Ca-plagioclase	2.4	2.3	Ca-plagioclase	4.2	2.8			
Lithic grain	ND		Lithic grain	ND		Lithic grain	14.5	5.2
Calcite	1.2	0.3	Calcite	2.9	1.6	Calcite	ND	
Dolomite	1.8	0.2	Dolomite	2.3	2.1	Dolomite	ND	
Muscovite	17.5	9.2	Muscovite	8.9	5.4	Micas	9.8	21.9
Biotite	10.4	4.3	Biotite	5.5	3.0			
Chlorite	2.8	1.4	Chlorite	1.7	1.1	Chlorite	ND	
Clay	19.5	18.6	Clay	9.8	3.6	Clay	ND	
Fe-oxides	0.2	0.2	Fe-oxides	0.1	0.1	Fe-oxides	ND	
Fibrolite	ND		Fibrolite	ND		Fibrolite	0.1	0.2
Opagues	1.3	0.1	Opagues	1.0	0.7	Opagues	1.1	2.8
Zircon	0.1	0.02	Zircon	0.1	0.3	Zircon	0.1	0.4
Tourmaline	0.2	0.1	Tourmaline	0.2	0.1	Tourmaline	0.3	0.7
Rutile	0.2	0.2	Rutile	0.2	0.2	Rutile	0.1	0.2
Ti oxide	0.4	0.1	Ti oxide	0.2	0.2	Ti oxide	0.0	0.1
Titanite	0.1	0.1	Titanite	0.2	0.3	Titanite	0.3	0.5
Apatite	0.1	0.04	Apatite	0.1	0.1	Apatite	0.1	0.4
Monazite	ND		Monazite	ND		Monazite	0.1	0.3
Amphibole	0.5	0.6	Amphibole	1.1	0.6	Amphibole	1.4	2.0
Pyroxene	0.2	0.1	Pyroxene	0.3	0.2	Pyroxene	0.3	0.5
Spinel	ND		Spinel	ND		Cr-spinel	ND	
Epidote	1.4	0.8	Epidote	1.0	0.3	Epidote	1.0	1.5
Allanite	ND		Allanite	0.02	0.1	Allanite	ND	
Chloritoid	ND		Chloritoid	ND		Chloritoid	ND	
Garnet	0.4	0.5	Garnet	1.2	1.9	Garnet	2.8	6.5
Staurolite	ND		Staurolite	0.03	0.02	Staurolite	0.1	0.1
Kyanite	0.04	0.1	Kyanite	0.1	0.1	Kyanite	0.2	0.5
Sillimanite	ND		Sillimanite	0.1	0.1	Sillimanite	0.02	0.1
<i>Sum</i>	<i>102.2</i>		<i>Sum</i>	<i>101.4</i>		<i>Sum</i>	<i>100.3</i>	

ND: not determined or under detection limit

Supplementary Table B: Major and Trace element concentrations in sediments from rivers draining monolithologic basins

Sample Name	R 94-12	MO 102	MO 112	PB 33	PB 37	MO 207	MO 73
Locality	Sarmi, Nepal	Confluence Darondi/Marsel Khola, Nepal	Confluence Bhuri Gandaki/Isul Khola, Nepal	Nepal	Nepal	Waling, Nepal	Nepal
Latitude	29.06	28.04111	28.04694	28.48966	28.43248	28.04288	28.25139
Longitude	82.50	84.66944	84.81472	83.65016	83.59952	83.79026	84.67278
River	Tributary of the Bheri River	Marsel Khola	Isul Khola	Ghara khola	Beg Khola	Andi Khola	Tributary of the Chepe khola
Sampling date	16-Mar-94	11-May-97	12-May-97	9-Jul-05	9-Jul-05	18-May-97	8-May-97
Drained lithology	Lesser Himalaya	Lesser Himalaya	Lesser Himalaya	Lesser Himalaya	Lesser Himalaya	Lesser Himalaya	High Himalayan Crystalline
Type of sediment	Bedload	Bank	Bank, gravel sample	Suspended Load	Bank	Bedload	Bank
Sampling Depth (m)				0			
ppm							
Cs	1.97	3.75	2.73	5.37	1.53	2.86	3.18
Rb	48.8	74.9	60.6	131	29.6	53.9	19.5
Ba	318	269	475	438	97.0	176	181
Th	3.14	6.91	15.6	11.3	8.45	9.77	8.41
U	1.05	1.64	3.13	2.82	1.93	2.30	1.53
Nb	2.72	9.83	7.89	13.0	9.61	8.19	10.4
Ta	0.281	0.987	0.893	0.986	0.933	0.837	0.872
La	8.83	19.7	55.3	25.5	30.9	27.7	13.1
Ce	17.6	50.5	113	66.5	65.7	66.7	50.8
Pr	1.97	4.36	12.1	6.21	7.25	6.31	3.62
Pb	87.1	12.4	9.19	17.1	11.6	9.12	8.94
Nd	7.08	15.6	42.9	23.5	27.5	22.3	14.1
Sr	55.0	53.0	41.1	53.8	34.3	27.4	52.1
Sm	1.22	2.90	7.66	4.86	5.41	4.05	2.99
Zr	39.1	168	268	216	220	227	146
Hf	0.982	4.39	6.62	5.58	5.22	5.76	3.68
Ti	649	2088	2810	5960	6246	2279	2584
Eu	0.223	0.431	1.28	1.06	1.15	0.653	0.581
Gd	1.04	2.21	5.84	4.29	4.42	3.10	2.71
Tb	0.145	0.313	0.865	0.667	0.658	0.472	0.446
Dy	0.928	1.74	5.42	4.22	3.92	2.92	3.02
Ho	0.181	0.333	1.10	0.872	0.751	0.614	0.683
Y	5.26	9.47	31.8	25.2	21.6	18.5	21.5
Er	0.516	0.943	3.27	2.51	2.06	1.90	2.26
Li	25.9	3.15	12.6	29.7	8.90	10.1	19.3
Yb	0.473	0.879	3.24	2.38	1.78	1.94	2.58
Lu	0.0718	0.128	0.463	0.355	0.253	0.281	0.397
Sc	2.53	3.6	8.79	17.8	10.4	4.81	3.60
V	15.4	35.1	34.6	168	65.1	30.3	114
Cr	14.7	43.0	25.0	111	33.5	21.9	52.2
Co	2.74	6.96	5.92	27.2	10.8	5.63	7.71
Ni	13.8	20.3	10.3	57.5	14.2	10.8	18.9
Cu	14.0	3.91	11.3	52.0	21.0	4.99	8.50
Zn	113	36.5	29.3	68.4	34.6	27.6	40.3
As	12.4	4.25	2.04	9.47	4.85	4.37	0.404
wt. %							
SiO2	22.24	81.82	79.89	54.44	83.90	78.27	76.81
Al2O3	2.78	8.82	6.36	18.12	5.59	6.04	10.69
Fe2O3	1.02	2.98	3.88	9.81	4.06	2.66	4.27
MnO	0.03	0.00	0.08	0.09	0.13	0.06	0.08
MgO	15.31	1.25	1.74	4.66	1.25	2.46	1.34
CaO	21.69	0.02	2.41	1.74	1.03	2.49	1.50
Na2O	0.04	0.82	0.68	1.25	1.13	0.16	1.20
K2O	1.33	1.96	1.40	3.46	0.64	2.19	1.93
TiO2	0.09	0.33	0.50	1.06	1.03	0.39	0.47
P2O5	0.10	0.08	0.11	0.15	0.10	0.13	0.12
LOI	35.10	1.80	2.80	5.65	1.30	5.15	1.53
Total	99.78	99.89	99.85	100.42	100.16	100.00	99.94

Sample Name	MO 50	MO 59	KN 83	MO 504	NAG 22	MAR-50	MAR-40
Locality	Nepal	Nepal	Ghyangphedi, Nepal	Tukche, Nepal	Pedi, Nepal	Temang, Nepal	Sabche, Nepal
Latitude	28.30056	28.29528	28.008	28.71120	28.78623	28.53000	28.63327
Longitude	84.63722	84.64639	85.454	83.64120	83.96370	84.31667	84.10613
River	Chepe khola	Chepe khola	Tadi khola	Yamkim khola	Marsyandi	Marsyandi	Sabche khola
Sampling date	6-May-97	7-May-97	21-Mar-94		28-Nov-95	15-Nov-00	12-Nov-00
Drained lithology	High Himalayan Crystalline	High Himalayan Crystalline	High Himalayan Crystalline	Tethys Himalaya	Tethys Himalaya	Tethys Himalaya	Tethys Himalaya
Type of sediment	Bank	Bank	Bedload	Suspended Load	Bedload	Bedload	Bedload
Sampling Depth (m)	0						
ppm							
Cs	4.16	6.21	4.24	9.58	30.5	7.72	7.52
Rb	56.9	99.6	101	180	144	113	118
Ba	310	524	452	553	633	351	447
Th	8.49	11.4	13.3	19.7	26.4	16.7	38.2
U	1.52	2.36	2.56	2.9	3.29	2.87	3.45
Nb	10.1	10.6	11.4	16.4	19.8	10.2	23.0
Ta	0.839	0.853	1.01	1.32	1.50	0.862	1.62
La	22.4	28.3	29.0	34.6	63.4	39.0	98.4
Ce	55.5	61.1	69.4	69.7	131	79.2	215
Pr	5.48	6.85	6.86	7.72	13.6	8.86	21.6
Pb	15.1	23.3	17.9	33.9	235	20.9	35.2
Nd	20.7	25.5	25.2	27.4	48.2	32.1	77.3
Sr	67.2	111	130	174	217	301	211
Sm	4.22	5.14	5.03	5.27	8.09	6.02	13.7
Zr	233	183	255	183	370	263	393
Hf	5.74	4.91	6.21	4.54	9.12	6.65	9.77
Ti	2385	2581	2588	3254	5117	2288	4862
Eu	0.856	0.948	0.966	1.00	1.42	0.971	2.19
Gd	4.22	4.70	4.39	4.71	6.15	5.12	10.1
Tb	0.824	0.888	0.694	0.733	0.889	0.764	1.48
Dy	6.61	6.28	4.52	4.49	5.28	4.45	8.25
Ho	1.55	1.45	0.963	0.911	1.01	0.888	1.60
Y	44.4	40.3	27.7	26.7	28.3	25.9	46.0
Er	4.94	4.38	2.94	2.66	2.98	2.59	4.55
Li	28.5	43.2	24.6	57.8	58.2	37.1	83.5
Yb	5.27	4.70	3.02	2.47	2.90	2.48	4.35
Lu	0.78	0.695	0.452	0.359	0.427	0.366	0.63
Sc	8.70	11.2	7.2	13.3	13.8	7.53	14.7
V	47.4	56.7	49.1	83.7	120	40.7	77.1
Cr	31.2	39.1	45.3	65.0	93.9	33.5	76.3
Co	7.41	8.45	7.56	10.6	16.5	7.54	19.6
Ni	12.6	15.9	18.9	26.3	30.5	15.6	55.3
Cu	10.8	13.9	17.7	16.0	12.5	8.59	30.9
Zn	53.0	59.6	52.2	85.6	179	36.6	81.9
As	7.84	10.8	0.712	9.26	20.5	21.3	11.3
wt. %							
SiO2	72.56	71.52	75.02	46.94	43.33	50.97	58.44
Al2O3	11.79	13.10	11.85	15.46	15.41	9.13	15.10
Fe2O3	6.04	5.36	3.76	5.14	5.60	2.95	6.13
MnO	0.27	0.19	0.07	0.05	0.04	0.03	0.04
MgO	1.01	1.48	1.05	2.66	2.10	1.58	0.61
CaO	1.18	1.35	1.39	11.19	13.07	17.24	6.24
Na2O	1.51	1.87	1.57	0.55	0.51	1.46	0.85
K2O	1.73	2.37	2.50	4.22	3.72	2.31	2.79
TiO2	0.42	0.47	0.45	0.56	0.80	0.38	0.73
P2O5	0.11	0.16	0.08	0.20	0.16	0.06	0.17
LOI	1.55	2.08	1.26	12.74	14.95	13.42	9.02
Total	98.17	99.95	98.99	99.71	99.69	99.53	100.12

Supplementary Table A: Major and Trace element concentrations in sediments from the Ganga and its major tributaries and the Marsyandi River

Sample Name	BR 931	BR 932	BR 922*	BR 924*	BR 943*	BR 946*	BR 946 Dup
Locality	Devrapayag, India	Devrapayag, India	Rishikesh, India	Rishikesh, India	Kanpur, India	Kanpur, India	Kanpur, India
Latitude			30.12661	30.12661	26.61306	26.61306	26.61306
Longitude			78.33044	78.33044	80.27576	80.27576	80.27576
River	Ganga	Ganga	Ganga	Ganga	Ganga	Ganga	Ganga
Sampling date	12-Aug-09	12-Aug-09	11-Aug-09	11-Aug-09	18-Aug-09	18-Aug-09	18-Aug-09
Type of sediment	Suspended Load	Suspended Load	Suspended Load	Bank	Suspended Load	Bedload	Bedload
Sampling Depth (m)	0	0	0.2		1.7		
Cs	16.4	8.00	14.6	8.47	10.8	5.61	4.44
Rb	185	115	170	128	125	136	68.4
Ba	469	344	436	368	391	328	221
Th	16.1	20.7	16.3	14.4	12.6	35.4	26.3
U	6.24	5.75	4.93	3.83	2.89	7.64	4.76
Nb	13.6	11.0	13.6	11.0	12.6	13.9	15.5
Ta	1.58	1.13	1.60	1.24	1.30	1.65	1.83
La	31.6	44.6	32.5	29.2	26.6	91.1	58.4
Ce	66.1	90.1	69.9	61.4	58.5	170	128
Pr	7.56	10.3	7.65	6.75	6.28	21.2	13.5
Pb	29.2	21.0	29.1	21.4	23.5	18.9	15.8
Nd	27.7	37.2	28.0	25.0	23.5	76.6	48.3
Sr	88.8	90.9	86.5	97.4	71.5	107	69.0
Sm	5.81	7.18	5.77	4.87	4.74	15.1	9.45
Zr	205	290	150	202	163	388	417
Hf	5.59	8.23	3.96	5.20	4.07	10.3	10.6
Ti	2750	2619	2836	2510	2841	3407	3675
Eu	0.916	0.972	0.942	0.813	0.891	1.94	1.16
Gd	5.22	5.87	5.14	4.16	4.26	12.1	8.19
Tb	0.868	0.888	0.789	0.653	0.678	1.98	1.39
Dy	5.44	5.06	4.73	3.96	4.25	12.0	9.20
Ho	1.10	1.00	0.926	0.784	0.846	2.42	1.94
Y	32.5	29.1	26.8	23.0	24.9	72.5	58.8
Er	3.16	2.90	2.62	2.25	2.47	7.05	5.88
Li	62.9	31.5	56.5	35.2	43.8	29.6	18.4
Yb	3.03	2.83	2.47	2.15	2.38	6.83	5.92
Lu	0.449	0.428	0.344	0.315	0.339	0.993	0.869
Sc	10.1	8.72	7.93	7.61	9.55	12.3	6.65
V	56.4	43.4	58.3	44.5	62.1	36.8	37.0
Cr	43.3	32.4	38.3	34.2	53.3	27.1	29.5
Co	9.52	7.66	10.0	6.87	10.6	5.01	4.94
Ni	22.5	14.7	22.5	16.3	29.5	7.70	9.82
Cu	19.7	14.4	33.6	17.2	25.3	2.86	6.15
Zn	56.4	41.9	70.7	46.0	68.9	31.1	39.8
As	15.8	9.01	13.5	7.43	11.9	3.00	3.22
wt. %							
SiO2	65.51	73.12	57.94	70.03	69.75	80.14	
Al2O3	12.28	9.37	12.26	9.95	12.09	8.32	
Fe2O3	4.01	3.19	4.40	3.15	4.51	3.59	
MnO	0.07	0.06	0.07	0.05	0.07	0.15	
MgO	2.99	2.24	4.98	2.64	2.09	1.01	
CaO	4.12	3.54	6.11	3.94	1.78	1.80	
Na2O	1.75	1.56	1.46	1.60	1.23	1.41	
K2O	3.25	2.27	3.20	2.57	2.82	1.51	
TiO2	0.48	0.44	0.50	0.42	0.50	0.63	
P2O5	0.13	0.12	0.14	0.11	0.14	0.23	
LOI	6.35	4.60		4.96		1.38	
Total	100.94	100.51	91.06	99.42	94.96	100.16	

* Major element concentrations already published in Lupker et al. (2012)

** Major element concentrations already published in Galy and France-Lanord (2001)

Sample Name	BGP 6**	BGP 6 Dup	BR 415*	BR 414*	BR 413*	BR 412*	BR 411*
Locality	Rajshahi, India	Rajshahi, India	Harding bridge, Bangladesh	Harding bridge, Bangladesh	Harding bridge, Bangladesh	Harding bridge, Bangladesh	Harding bridge, Bangladesh
Latitude	24.35833	24.35833	24.05290	24.05290	24.05290	24.05290	24.05290
Longitude	88.60833	88.60833	89.02465	89.02465	89.02465	89.02465	89.02465
River	Ganga	Ganga	Ganga	Ganga	Ganga	Ganga	Ganga
Sampling date	2-Aug-93	2-Aug-93	13-Jul-04	13-Jul-04	13-Jul-04	13-Jul-04	13-Jul-04
Type of sediment	Bedload	Bedload	Suspended Load	Suspended Load	Suspended Load	Suspended Load	Suspended Load
Sampling Depth (m)			0	2	4	6.5	9
Cs	3.83	4.02	15.4	13.6	11.8	11.7	8.08
Rb	77.9	85.2	202	186	174	175	107
Ba	294	307	627	562	531	527	372
Th	30.2	31.3	20.0	19.0	19.3	18.0	15.1
U	5.41	5.71	4.64	4.08	4.15	3.96	2.92
Nb	12.9	12.0	17.4	17.9	16.5	15.7	13.6
Ta	1.78	1.44	1.54	1.57	1.45	1.39	1.37
La	67.1	70.2	42.0	41.3	42.2	40.0	32.8
Ce	144	145	89.4	88.1	89.2	84.7	76.3
Pr	15.5	16.4	9.75	9.77	9.87	9.29	7.84
Pb	17.8	17.6	30.4	26.2	25.3	25.3	17.4
Nd	56.5	60.8	35.0	35.7	35.8	34.0	29.0
Sr	115	117	84.1	91.0	99.3	101	88.6
Sm	10.9	11.4	6.93	6.98	7.01	6.77	5.60
Zr	683	667	178	204	227	199	249
Hf	17.1	17.4	4.73	5.22	5.79	5.17	6.14
Ti	3155	3166	4137	4198	3976	3833	3314
Eu	1.39	1.45	1.27	1.26	1.23	1.20	1.04
Gd	8.51	9.10	6.19	6.15	6.05	5.80	4.96
Tb	1.25	1.29	0.984	0.945	0.93	0.872	0.782
Dy	7.24	7.61	5.97	5.74	5.72	5.42	4.81
Ho	1.39	1.48	1.17	1.15	1.13	1.06	0.95
Y	41.1	43.5	32.7	32.8	32.5	30.9	27.1
Er	3.99	4.24	3.33	3.30	3.21	3.07	2.65
Li	14.7	16.3	51.8	48.2	43.5	41.9	32.2
Yb	3.87	4.16	3.18	3.11	3.06	2.88	2.51
Lu	0.587	0.614	0.461	0.45	0.448	0.417	0.366
Sc	7.74	9.81	15.0	14.4	13.5	12.8	9.04
V	32.7	34.8	93.4	90.7	81.1	79.1	61.8
Cr	36.9	38.4	76.6	81.9	67.0	63.4	53.4
Co	3.93	4.32	15.5	15.1	13.6	13.4	10.3
Ni	11.0	11.3	38.6	40.5	32.4	31.5	26.4
Cu	10.9	11.3	33.9	31.7	25.7	23.7	24.3
Zn	29.1	28.1	84.9	83.5	73.3	72.1	56.6
As	2.32	5.03	9.17	7.33	5.52	5.70	5.41
wt. %							
SiO2	78.45		57.76	60.42	63.07	63.44	68.36
Al2O3	7.86		18.57	17.04	15.64	15.06	12.27
Fe2O3	2.46		6.61	6.31	5.75	5.55	4.20
MnO	0.05		0.08	0.08	0.07	0.07	0.06
MgO	0.91		2.39	2.34	2.20	2.07	1.71
CaO	3.28		1.79	2.36	2.73	2.67	2.77
Na2O	1.48		1.03	1.13	1.19	1.22	1.37
K2O	1.83		3.77	3.52	3.28	3.20	2.82
TiO2	0.57		0.75	0.76	0.72	0.70	0.58
P2O5	0.12		0.16	0.15	0.15	0.14	0.12
LOI	2.57		7.19	6.51	6.07	6.13	4.66
Total	99.57		100.09	100.62	100.87	100.23	98.89

Sample Name	BR 418*	BR 8252*	BR 8253*	BR 717*	PB 79*	PB 80*	PB 80 Dup
Locality	Harding bridge, Bangladesh	Harding bridge, Bangladesh	Harding bridge, Bangladesh	Harding bridge, Bangladesh	Chisapani, Nepal	Chisapani, Nepal	Chisapani, Nepal
Latitude	24.05290	24.05290	24.05290	24.05290	28.63895	28.63895	28.63895
Longitude	89.02465	89.02465	89.02465	89.02465	81.28278	81.28278	81.28278
River	Ganga	Ganga	Ganga	Ganga	Kamali	Kamali	Kamali
Sampling date	13-Jul-04	10-Sep-08	10-Sep-08	17-Aug-07	15-Jul-05	15-Jul-05	15-Jul-05
Type of sediment	Bedload	Bedload	Suspended Load	Bedload	Suspended Load	Bank	Bank
Sampling Depth (m)	10	14	0	11	0		
Cs	2.64	3.05	8.92	3.87	8.20	3.50	3.56
Rb	56.6	34.5	51.7	50.3	141	70.7	71.2
Ba	212	203	240	244	482	255	262
Th	29.7	24.3	16.1	74.2	19.0	12.0	12.3
U	5.14	3.33	1.85	11.7	3.33	2.92	3.07
Nb	18.4	13.6	15.8	20.3	16.9	9.79	9.79
Ta	2.08	2.91	1.31	2.50	1.57	1.04	1.01
La	71.0	41.6	30.8	141	39.8	27.9	27.9
Ce	146	110	75.4	292	82.0	59.7	59.2
Pr	16.7	10.6	8.12	33.1	8.78	6.52	6.55
Pb	15.0	10.7	10.5	14.2	12.7	13.9	14.0
Nd	60.4	39.4	31.2	121	31.6	23.7	23.9
Sr	103	83.6	56.5	98.3	96.3	93.8	94.5
Sm	12.0	7.63	6.27	22.3	5.69	4.55	4.70
Zr	479	403	196	1550	259	285	337
Hf	12.1	10.5	5.13	40.5	6.43	7.03	8.49
Ti	4355	2523	4323	5299	3606	2395	2485
Eu	1.48	0.928	1.19	2.24	0.927	0.74	0.817
Gd	10.1	6.41	5.44	17.7	4.37	3.90	4.38
Tb	1.64	0.977	0.854	2.52	0.644	0.603	0.746
Dy	10.3	5.70	5.32	14.7	3.90	3.84	5.02
Ho	2.12	1.16	1.06	2.83	0.787	0.776	1.02
Y	62.0	34.5	31.1	85.6	23.0	22.6	30.5
Er	6.35	3.33	3.04	8.11	2.26	2.24	2.97
Li	15.8	12.6	45.3	15.7	28.1	16.9	17.0
Yb	6.23	3.32	2.95	8.17	2.18	2.28	2.79
Lu	0.905	0.50	0.433	1.25	0.312	0.333	0.416
Sc	13.2	4.56	11.8	14.3	10.7	6.63	6.95
V	46.9	34.5	100	53.5	60.6	33.0	32.4
Cr	40.0	28.2	73.6	44.1	47.2	23.2	22.3
Co	5.58	4.46	13.8	6.40	4.77	5.15	4.89
Ni	11.7	9.39	39.4	12.4	12.4	11.0	11.0
Cu	10.0	7.82	39.4	7.10	6.19	7.33	7.38
Zn	38.8	24.2	70.4	31.0	30.2	29.2	29.1
As	1.79	2.21	14.6	2.63	6.24	3.80	3.85
wt. %							
SiO2	75.40	80.96	56.94	75.03	59.03	77.77	
Al2O3	8.58	7.68	15.64	8.31	10.57	6.50	
Fe2O3	5.19	2.57	6.31	4.53	3.94	2.57	
MnO	0.19	0.06	0.09	0.13	0.06	0.05	
MgO	1.47	0.90	2.74	1.17	3.46	1.42	
CaO	4.15	2.50	4.25	3.29	8.69	4.56	
Na2O	1.18	1.41	0.97	1.29	1.15	0.92	
K2O	1.26	1.74	3.28	1.72	2.75	1.56	
TiO2	0.91	0.47	0.75	0.90	0.51	0.45	
P2O5	0.27	0.10	0.14	0.19	0.12	0.10	
LOI	2.15	2.19	8.93	2.65	10.51	4.58	
Total	100.73	100.58	100.04	99.18	100.79	100.49	

Sample Name	BR 8113*	BR 8113 Dup	BR 8115*	LO 762*	LO 763*	LO 763 Dup	PB 69*
Locality	Revelganj, India	Revelganj, India	Revelganj, India	Chatara, Nepal	Chatara, Nepal	Chatara, Nepal	Chatara, Nepal
Latitude	25.77950	25.77950	25.77950	26.84815	26.84815	26.84815	26.84815
Longitude	84.63070	84.63070	84.63070	87.15137	87.15137	87.15137	87.15137
River	Kamali	Kamali	Kamali	Kosi	Kosi	Kosi	Kosi
Sampling date	18-Aug-08	18-Aug-08	18-Aug-08	16-Aug-07	16-Aug-07	16-Aug-07	13-Jul-05
Type of sediment	Suspended Load	Suspended Load	Bedload	Suspended Load	Bank	Bank	Suspended Load
Sampling Depth (m)	0	0	5.4	0			0
Cs	11.5	11.7	4.31	11.6	5.26	5.84	13.8
Rb	173	176	101	107	77.2	106	177
Ba	574	586	305	357	290	341	637
Th	18.5	18.4	8.06	19.5	12.1	11.8	21.0
U	3.42	3.47	1.72	4.23	2.15	3.08	4.06
Nb	15.2	15.2	5.94	17.9	9.23	9.48	18.9
Ta	1.28	1.27	0.554	1.63	0.878	0.902	1.73
La	39.9	40.5	21.5	40.9	28.3	22.3	48.0
Ce	86.0	86.3	36.5	101	61.7	68.5	109
Pr	9.43	9.53	4.55	9.92	6.70	4.86	11.2
Pb	30.3	30.5	16.0	17.8	17.6	19.1	17.3
Nd	34.7	34.7	16.8	36.4	24.3	17.7	40.0
Sr	93.5	93.8	86.3	82.2	74.2	88.7	61.9
Sm	6.72	6.65	3.07	7.25	4.80	3.28	7.60
Zr	186	195	112	286	203	206	171
Hf	4.81	5.07	3.20	7.79	5.17	5.30	4.62
Ti	3763	3708	1382	4017	2013	1961	3979
Eu	1.23	1.23	0.576	1.17	0.872	0.601	1.24
Gd	5.78	5.85	2.70	6.15	4.21	2.88	6.03
Tb	0.88	0.891	0.376	0.92	0.658	0.447	0.875
Dy	5.33	5.43	2.40	5.45	4.03	2.76	5.00
Ho	1.05	1.07	0.468	1.06	0.79	0.571	0.969
Y	30.3	30.3	13.7	30.8	23.2	18.2	27.2
Er	2.99	3.03	1.32	3.02	2.29	1.73	2.73
Li	46.5	46.7	17.0	47.9	17.2	1.23	44.0
Yb	2.85	2.89	1.27	2.94	2.18	1.67	2.60
Lu	0.413	0.413	0.179	0.426	0.305	0.25	0.374
Sc	12.1	13.0	6.08	8.04	4.90	3.49	11.3
V	87.1	86.7	24.8	73.2	33.7	33.7	83.9
Cr	59.3	61.9	18.6	52.1	29.9	29.1	65.2
Co	13.2	13.8	4.25	13.2	6.20	6.10	13.3
Ni	30.5	31.0	9.55	28.6	40.4	16.3	32.2
Cu	26.8	27.1	2.72	26.4	50.2	13.2	20.2
Zn	71.9	78.6	20.5	83.2	52.8	38.5	59.6
As	11.4	11.5	3.24	9.01	0.434	0.651	6.17
wt. %							
SiO2	56.24		79.56	64.64	77.58		57.44
Al2O3	14.83		7.03	14.86	9.87		17.54
Fe2O3	5.75		1.72	5.34	2.92		6.00
MnO	0.08		0.03	0.07	0.04		0.07
MgO	3.26		1.17	2.15	1.20		3.78
CaO	4.80		3.41	1.88	1.58		2.24
Na2O	0.87		1.22	1.78	1.72		1.59
K2O	3.50		1.87	3.61	2.35		4.52
TiO2	0.65		0.24	0.68	0.38		0.73
P2O5	0.13		0.06	0.17	0.11		0.15
LOI	9.56		3.79	3.80	1.95		5.83
Total	99.67		100.08	98.98	99.68		99.91

Sample Name	PB 70*	BR 331*	BR 332*	LO 757*	LO 756*	LO 754*	LO 755*
Locality	Chatara, Nepal	Dumarighat, India	Dumarighat, India	Narayanghat, Nepal	Narayanghat, Nepal	Narayanghat, Nepal	Narayanghat, Nepal
Latitude	26.84815	25.54113	25.54113	27.70297	27.70297	27.70297	27.70297
Longitude	87.15137	86.72231	86.72231	84.42661	84.42661	84.42661	84.42661
River	Kosi	Kosi	Kosi	Narayani	Narayani	Narayani	Narayani
Sampling date	13-Jul-05	10-May-04	10-May-04	12-Aug-07	12-Aug-07	12-Aug-07	12-Aug-07
Type of sediment	Bank	Bank	Bank	Suspended Load	Suspended Load	Suspended Load	Suspended Load
Sampling Depth (m)				0	3.3	6	8
Cs	4.77	11.1	8.43	10.9	10.6	10.3	10.6
Rb	91.8	107	101	146	142	141	142
Ba	294	362	334	460	454	437	441
Th	14.7	19.4	13.6	14.8	15.2	15.8	15.5
U	2.75	3.79	2.60	3.18	4.26	3.33	3.26
Nb	9.21	19.4	13.5	13.6	13.1	12.4	12.6
Ta	1.08	1.75	1.33	1.25	1.33	1.24	1.23
La	32.4	40.9	29.7	32.8	35.8	36.9	35.4
Ce	71.1	97.8	73.5	68.7	73.9	77.1	73.7
Pr	7.59	10.0	7.14	7.43	8.07	8.48	8.06
Pb	15.3	15.7	17.2	24.8	72.0	23.6	24.1
Nd	27.8	36.9	26.7	26.9	29.6	30.9	29.7
Sr	92.6	60.1	67.2	234	213	197	194
Sm	5.43	7.43	5.30	5.26	5.74	6.03	5.82
Zr	159	281	202	217	220	213	200
Hf	3.96	7.57	5.30	5.54	5.63	6.34	5.18
Ti	1903	4248	2884	3021	2916	2710	2777
Eu	0.871	1.24	0.903	0.984	0.973	1.03	0.997
Gd	4.59	6.39	4.59	4.52	4.94	5.05	4.81
Tb	0.724	0.974	0.714	0.712	0.706	0.74	0.734
Dy	4.54	5.93	4.11	4.20	4.40	4.29	4.44
Ho	0.922	1.18	0.815	0.838	0.843	0.815	0.875
Y	27.2	34.9	24.0	25.1	24.0	24.1	25.1
Er	2.76	3.45	2.31	2.47	2.43	2.29	2.52
Li	17.6	40.3	27.1	45.9	43.9	41.2	43.7
Yb	2.65	3.27	2.26	2.36	2.38	2.18	2.34
Lu	0.386	0.476	0.326	0.343	0.35	0.323	0.344
Sc	5.51	9.96	5.14	9.72	9.31	8.90	9.24
V	29.2	78.0	51.8	57.8	55.0	52.0	53.3
Cr	24.8	61.7	37.0	48.0	44.6	41.9	44.0
Co	5.82	13.9	9.26	9.07	8.94	8.28	8.37
Ni	11.9	36.3	20.9	21.5	20.0	18.9	20.1
Cu	9.31	37.6	34.0	17.6	14.7	16.2	14.9
Zn	32.7	77.9	56.5	68.2	64.9	60.8	63.4
As	3.07	11.3	4.04	11.0	11.9	9.86	9.82
wt. %							
SiO2	79.11	61.28	72.72	53.31	56.78	59.47	57.77
Al2O3	9.13	16.30	12.31	11.17	11.34	10.97	11.17
Fe2O3	2.96	5.96	4.11	3.79	3.82	3.60	3.66
MnO	0.06	0.08	0.06	0.05	0.05	0.05	0.05
MgO	1.26	2.46	1.36	2.19	2.07	2.00	2.05
CaO	1.82	2.29	1.39	11.86	10.91	9.38	9.70
Na2O	1.60	1.31	1.58	1.36	1.38	1.47	1.45
K2O	1.95	3.96	2.95	2.78	2.84	2.72	2.78
TiO2	0.34	0.74	0.49	0.49	0.49	0.46	0.47
P2O5	0.13	0.16	0.13	0.11	0.11	0.11	0.11
LOI	1.70	4.93	2.55	11.64	10.10	8.93	9.40
Total	100.05	99.46	99.63	98.74	99.89	99.16	98.60

Sample Name	LO 755 Dup	LO 758 C*	PB 54*	LO 1002	LO 1007	LO 1007 Dup	BR 8106*
Locality	Narayanghat, Nepal	Narayanghat, Nepal	Narayanghat, Nepal	Narayanghat, Nepal	Narayanghat, Nepal	Narayanghat, Nepal	Hajipur, India
Latitude	27.70297	27.70297	27.70297				25.67933
Longitude	84.42661	84.42661	84.42661				85.19362
River	Narayani	Narayani	Narayani	Narayani	Narayani	Narayani	Narayani
Sampling date	12-Aug-07	12-Aug-07	11-Jul-05	16-Jul-10	16-Jul-10	16-Jul-10	17-Aug-08
Type of sediment	Suspended Load	Suspended Load	Suspended Load	Suspended Load	Suspended Load	Suspended Load	Suspended Load
Sampling Depth (m)	8	8.5	8	0	0	0	0
Cs	10.8	5.02	8.5	12.7	3.43	3.14	11.1
Rb	148	85.3	123	170	70.4	62.0	141
Ba	457	290	437	510	307	280	497
Th	14.9	7.87	11.7	14.4	13.4	12.3	17.2
U	3.24	2.02	2.69	3.38	3.33	2.96	3.20
Nb	13.2	6.79	10.7	13.2	14.5	12.9	15.2
Ta	1.30	0.581	1.01	1.18	1.06	0.922	1.28
La	33.2	22.2	28.9	32.1	37.5	34.0	38.3
Ce	69.1	45.8	60.2	66.2	79.9	72.8	81.8
Pr	7.57	4.95	6.7	7.43	8.82	7.99	8.9
Pb	25.0	15.5	19.3	27.4	13.6	12.2	26.7
Nd	27.3	17.8	24.2	27.3	32.9	29.5	32.4
Sr	200	112	177	269	68.9	63.0	159
Sm	5.41	3.41	4.70	5.22	6.22	5.45	6.21
Zr	236	113	188	187	209	190	190
Hf	5.88	2.98	4.83	4.85	5.30	4.74	5.00
Ti	2840	1519	2568	3291	2542	2272	3532
Eu	0.986	0.64	0.894	0.97	1.09	0.98	1.16
Gd	4.68	2.86	4.10	4.34	5.00	4.45	5.34
Tb	0.728	0.423	0.627	0.686	0.774	0.675	0.825
Dy	4.53	2.57	3.87	4.15	4.61	4.23	4.88
Ho	0.913	0.532	0.786	0.819	0.968	0.853	0.966
Y	26.9	15.7	22.6	24.0	28.2	25.3	28.0
Er	2.65	1.61	2.25	2.42	2.83	2.56	2.76
Li	46.7	22.3	32.1	51.6	14.8	13.4	45.3
Yb	2.53	1.56	2.17	2.27	2.79	2.50	2.63
Lu	0.371	0.226	0.307	0.334	0.402	0.363	0.381
Sc	9.38	5.78	8.52	10.5	5.00	5.26	10.6
V	54.9	36.1	51.9	69.4	46.6	41.9	79.9
Cr	45.6	38.3	38.8	58.9	32.0	28.7	59.5
Co	8.55	4.54	7.64	11.2	7.43	6.54	13.4
Ni	20.7	18.7	16.7	28.7	17.8	17.3	32.6
Cu	16.5	11.9	10.1	25.8	25.0	22.2	112
Zn	65.0	37.8	49.7	80.8	42.8	38.2	128
As	9.57	1.68	5.12	14.4	2.84	2.52	12.3
wt. %							
SiO2		69.39	62.71	53.70	75.74		53.31
Al2O3		8.44	10.20	12.22	8.21		14.51
Fe2O3		2.55	3.63	4.60	3.62		5.32
MnO		0.05	0.06	0.06	0.08		0.07
MgO		2.49	2.31	2.50	2.23		2.86
CaO		5.90	8.83	12.36	2.92		8.13
Na2O		1.36	1.45	1.34	1.33		1.23
K2O		1.97	2.61	3.19	1.54		3.57
TiO2		0.28	0.45	0.57	0.44		0.61
P2O5		0.07	0.11	0.13	0.14		0.14
LOI		6.40	8.19		3.73		10.28
Total		98.91	100.55	90.65	99.97		100.03

Sample Name	BR 8107*	MAR-55	MAR-58	MAR-70	MAR-31	MAR-29	MAR-19
Locality	Hajipur, India	Tal, Nepal	Chamje, Nepal	Besi Sahar, Nepal	Philesangu, Nepal	Suribar, Nepal	Turture, Nepal
Latitude	25.67933	28.46715	28.44050	28.23178	28.19050	28.13133	28.03650
Longitude	85.19362	84.37253	84.38800	84.38290	84.41967	84.44050	84.46467
River	Narayani	Marsyandi	Marsyandi	Marsyandi	Marsyandi	Marsyandi	Marsyandi
Sampling date	17-Aug-08	16-Nov-00	16-Nov-00	18-Nov-00	6-Nov-00	6-Nov-00	4-Nov-00
Type of sediment	Bedload	Bedload	Bedload	Bedload	Bedload	Bedload	Bedload
Sampling Depth (m)							
Cs	7.48	17.5	14.4	6.32	5.80	7.30	6.07
Rb	111	194	166	94.6	78	104	84.2
Ba	390	420	283	328	284	331	279
Th	13.6	16.0	16.3	46.3	26.8	8.24	21.0
U	2.39	8.37	5.68	8.87	5.58	5.64	5.12
Nb	9.14	8.99	12.8	26.5	11.5	9.84	11.7
Ta	0.844	0.986	1.79	3.6	1.17	0.865	1.61
La	30.9	37.5	38.1	97.1	68.2	22.2	50.7
Ce	68.6	60.6	79.0	201	134	44.8	104
Pr	7.11	8.85	8.99	22.9	15.4	5.10	11.6
Pb	20.9	29.5	28.4	20.4	17.7	17.1	17.8
Nd	25.5	32.7	33.4	84.5	56.5	18.8	43.6
Sr	128	276	220	120	146	135	133
Sm	4.82	7.63	7.11	16.7	10.9	3.75	8.60
Zr	189	137	160	154	196	123	159
Hf	4.78	3.73	4.41	4.33	5.13	3.32	4.41
Ti	2085	1536	1948	4254	2472	2178	2494
Eu	0.825	1.48	1.04	2.25	1.61	0.771	1.27
Gd	3.93	8.11	6.04	14.1	8.99	3.59	7.34
Tb	0.579	1.57	0.981	2.28	1.31	0.614	1.23
Dy	3.19	10.9	5.41	15.0	7.95	4.63	8.10
Ho	0.605	2.52	1.01	3.13	1.56	1.03	1.79
Y	17.5	77.2	28.3	91.6	44.8	29.5	50.9
Er	1.71	7.94	2.65	9.43	4.64	3.21	5.49
Li	25.1	72.3	52.5	27.0	24.0	30.8	24.6
Yb	1.64	7.58	2.40	9.16	4.54	3.31	5.61
Lu	0.239	1.09	0.336	1.29	0.671	0.481	0.812
Sc	6.21	7.84	5.99	15.3	13.4	10.2	11.4
V	44.6	26.8	29.5	66.7	43.6	49.1	45.0
Cr	41.0	35.1	25.2	53.7	41.6	39.7	36.6
Co	8.39	4.40	6.22	14.9	9.08	8.44	10.6
Ni	22.4	9.98	12.5	24.1	16.5	17.0	15.7
Cu	12.0	5.83	8.38	23.4	11.7	10.9	14.0
Zn	44.4	36.3	41.4	45.7	42.0	46.2	40.2
As	2.75	8.92	23.5	52.6	10.2	3.90	14.2
wt. %							
SiO ₂	71.09	60.58	61.26	64.74	69.63	70.04	71.31
Al ₂ O ₃	10.07	11.20	10.64	11.77	10.22	10.42	10.30
Fe ₂ O ₃	3.22	2.11	2.74	7.68	4.17	4.13	4.81
MnO	0.05		0.03	0.21	0.11	0.09	0.14
MgO	1.54	1.17	1.39	2.12	1.71	1.88	1.61
CaO	4.25	10.68	11.14	5.55	6.51	5.36	5.26
Na ₂ O	1.57	2.35	2.36	1.47	1.66	1.62	1.68
K ₂ O	2.55	3.13	2.74	1.98	1.58	2.03	1.64
TiO ₂	0.38	0.26	0.34	0.73	0.45	0.36	0.45
P ₂ O ₅	0.07	0.11	0.15	0.22	0.18	0.11	0.16
LOI	4.19	8.23	7.04	3.37	3.81	3.78	3.10
Total	98.98	99.82	99.83	99.84	100.03	99.82	100.46

Sample Name	MAR-12	MAR-10	MAR-10 Dup
Locality	Anpu, Nepal	Lobrang, Nepal	Lobrang, Nepal
Latitude	27.90783	27.86983	27.86983
Longitude	84.54217	84.55083	84.55083
River	Marsyandi	Marsyandi	Marsyandi
Sampling date	3-Nov-00	3-Nov-00	3-Nov-00
Type of sediment	Bedload	Bedload	Bedload
Sampling Depth (m)			
Cs	8.10	6.85	7.27
Rb	148	106	119
Ba	350	333	366
Th	11.5	10.2	10.7
U	2.87	2.06	2.29
Nb	8.02	8.17	8.85
Ta	0.932	0.738	0.793
La	25.0	27.4	31.8
Ce	49.4	47.6	53.3
Pr	5.41	5.78	6.53
Pb	23.6	18.1	18.9
Nd	20.3	20.9	23.4
Sr	155	146	151
Sm	5.04	4.00	4.22
Zr	141	109	56.0
Hf	3.69	2.95	1.62
Ti	1624	1765	1766
Eu	0.817	0.814	0.857
Gd	6.49	3.57	3.84
Tb	1.68	0.619	0.61
Dy	16.8	4.33	4.13
Ho	4.78	0.907	0.889
Y	137	25.5	25.1
Er	18.2	2.75	2.66
Li	28.9	27.8	27.4
Yb	20.2	2.67	2.62
Lu	2.68	0.382	0.38
Sc	9.45	13.8	10.4
V	34.6	38.4	37.4
Cr	30.6	39.0	34.7
Co	5.40	5.93	5.56
Ni	11.3	11.8	11.5
Cu	6.42	7.37	8.09
Zn	32.6	38.7	37.4
As	5.55	3.34	3.17
wt. %			
SiO ₂	71.18	71.15	
Al ₂ O ₃	9.93	9.60	
Fe ₂ O ₃	2.85	2.91	
MnO	0.05	0.05	
MgO	1.19	1.49	
CaO	5.86	6.04	
Na ₂ O	1.81	1.64	
K ₂ O	2.01	1.87	
TiO ₂	0.29	0.29	
P ₂ O ₅	0.11	0.14	
LOI	4.53	4.63	
Total	99.81	99.81	

Supplementary Table C: Trace-element concentrations in rock standards analyzed as unknown samples (ppm)

	BR ref		AGV-1 n=2	2 σ (%)	AGV-1		BEN n=10	2 σ (%)	BEN		JSd-2 n=28	2 σ (%)	JSd-2	
	Chauvel et al. (2011)	Chauvel et al. (2011)			Chauvel et al. (2011)	Diff (%)			Chauvel et al. (2011)	Diff (%)			Chauvel et al. (2011)	Diff (%)
Li	15.1	7.15	10.8	9.7	10.9	1.1	13.2	4.1	13.0	-1.2	22.8	5.4	22.1	-3.1
Sc	22.8	25.9	12.2	11.1	12.8	4.5	23.0	6.8	22.5	-2.3	17.8	6.2	17.9	0.8
Ti	15300	17500	5955	5.7	6080	2.0	15492	5.2	15400	-0.6	3569	4.0	3570	0.0
V	226	267	115	1.9	117	1.8	229	5.3	226	-1.4	125	4.4	126	0.9
Cr	398	425	7.91	4.5	8.05	1.7	348	5.7	340	-1.6	104	6.5	106	2.0
Co	56.6	52.6	14.9	0.9	15.3	2.3	61.0	5.7	60.0	-1.6	47.6	5.1	47.5	-0.2
Ni	259	238	14.6	2.8	14.5	-0.7	265	6.4	258	-2.8	91.7	6.2	91.1	-0.7
Cu	68.8	37.0	57.1	5.4	57.2	0.2	68.8	8.9	69.3	0.7	1040	9.0	1060	1.9
Zn	158	109	77.4	6.2	91.7	15.6	122	4.8	119	-2.6	1996	17.3	1980	-0.8
As	2.22		1.03	11.6	0.98	-5.1	2.23	37.6	1.80	-24.1	38.3	45.4	37.8	-1.4
Rb	46.6	80.0	68.0	9.5	66.8	-1.8	47.5	6.1	46.8	-1.6	26.3	8.2	25.8	-2.0
Sr	1340	607	662	1.9	657	-0.8	1381	3.5	1380	-0.1	213	7.6	210	-1.4
Y	28.9	28.5	19.4	1.4	19.5	0.6	29.3	4.4	29.0	-1.2	18.4	2.7	18.2	-1.3
Zr	273	294	237	1.0	237	0.2	278	4.4	274	-1.5	108	7.9	103	-4.7
Nb	115	38.1	14.5	0.2	13.9	-4.6	116	5.0	116	-0.2	4.39	8.3	4.19	-4.8
Cs	0.82	0.648	1.29	2.3	1.29	0.3	0.754	6.0	0.752	-0.2	1.07	3.9	1.05	-2.0
Ba	1090	390	1217	2.7	1230	1.0	1055	2.6	1050	-0.5	1264	4.4	1230	-2.7
La	82.6	33.6	38.3	0.5	38.0	-0.9	83.4	2.1	83.0	-0.5	11.3	4.0	10.8	-4.3
Ce	155	73.9	69.7	1.5	69.0	-1.0	157	2.8	155	-1.2	23.5	7.6	22.1	-6.5
Pr	17.4	9.61	8.39	0.5	8.47	0.9	17.6	3.4	17.4	-1.4	3.03	3.6	2.94	-3.0
Nd	66.4	39.9	31.7	0.5	31.8	0.2	67.2	3.1	67.0	-0.3	12.5	3.5	12.1	-3.3
Sm	12.2	8.36	5.75	4.7	5.77	0.3	12.3	3.5	12.2	-0.8	2.95	3.8	2.86	-3.2
Eu	3.66	2.53	1.61	4.2	1.61	0.3	3.67	3.0	3.67	0.0	0.853	5.8	0.872	2.2
Gd	9.88	7.28	4.68	0.0	4.69	0.1	9.83	2.5	9.87	0.5	3.04	3.7	2.85	-6.5
Tb	1.25	1.03	0.611	0.9	0.632	3.4	1.26	3.0	1.26	0.2	0.479	3.6	0.465	-3.0
Dy	6.42	5.77	3.57	5.1	3.55	-0.7	6.52	2.3	6.44	-1.2	3.10	3.3	3.04	-1.8
Ho	1.08	1.05	0.674	2.0	0.675	0.1	1.09	3.2	1.08	-1.1	0.651	2.5	0.641	-1.5
Er	2.58	2.70	1.85	5.6	1.84	-0.5	2.60	4.3	2.59	-0.4	1.95	2.7	1.91	-1.9
Yb	1.84	2.13	1.66	6.6	1.65	-0.8	1.86	3.8	1.84	-1.2	1.94	3.3	1.91	-1.8
Lu	0.246	0.297	0.247	4.6	0.241	-2.6	0.248	5.9	0.245	-1.3	0.293	4.8	0.288	-1.8
Hf	5.67	6.75	5.09	1.1	5.05	-0.8	5.72	3.2	5.64	-1.5	2.72	8.8	2.74	0.7
Ta	5.55	2.34	0.809	1.9	0.825	2.0	5.83	12.7	5.57	-4.6	0.369	17.7	0.345	-7.0
Pb	4.77	3.69	35.2	0.8	35.9	1.9	4.22	8.7	4.17	-1.2	160	7.2	154	-4.2
Th	10.7	4.75	6.37	2.8	6.35	-0.3	10.8	3.0	10.7	-0.9	2.62	3.3	2.43	-8.0
U	2.39	1.20	1.80	4.0	1.83	1.5	2.42	5.1	2.39	-1.1	1.13	10.2	1.09	-4.0

Standard deviations (2 σ) were calculated between the n measurement compositions of the rock standards analysed as unknown samples and calibrated to the BR or BR 24 standards using the reference values of Chauvel et al. (2011). Differences between the published values and our measurement compositions (Diff) are also reported.

Supplementary Table D: Present-day ratios used to calculate the present-day compositions of the reservoirs formed at different times in the past

	Nd (ppm)	¹⁴⁷ Sm/ ¹⁴⁴ Nd	¹⁴³ Nd/ ¹⁴⁴ Nd	Hf (ppm)	¹⁷⁶ Lu/ ¹⁷⁷ Hf	¹⁷⁶ Hf/ ¹⁷⁷ Hf	Sources in the literature
CHUR		0.1960	0.512630		0.0336	0.282785	Bouvier et al. (2008)
MORB	12.03	0.2107	0.513074	2.79	0.0383	0.283199	Gale et al. (2013) for Nd and Hf concentrations and for Nd isotopic composition. εHf calculated to be on the present-day Terrestrial Array of Vervoort et al. (2011)
Depleted Mantle	1.2		0.513074	0.3		0.283199	Gale et al. (2013) for Nd isotopic composition. εHf calculated to be on the present-day Terrestrial Array of Vervoort et al. (2011). Nd and Hf concentrations estimated at 10% of the MORB concentrations.
OIB		0.1374	0.512896		0.0082	0.282973	GEOROC database for Nd isotopic composition. εHf calculated to be on the present-day Terrestrial Array of Vervoort et al. (2011).
Oceanic sediment	28.0	0.1314	0.512180	1.78	0.0191	0.282829	Chauvel et al. (2008) for Hf isotopic composition. Plank (2013) for Nd and Hf concentrations and Nd isotopic compositions
Continental Crust		0.1179			0.0115		Rudnick and Gao (2003)

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