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The dependence of meteoric ^{10}Be concentrations on particle size in Amazon River bed sediment and the extraction of reactive $^{10}\text{Be}/^9\text{Be}$ ratios

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Abstract

Concentrations of meteoric ^{10}Be extracted from detrital Amazon river sediment are strongly particle size-dependent. Such grain size dependency presents a formidable obstacle to routine applications of meteoric cosmogenic ^{10}Be to basin-wide erosion studies. In this study, we explore means to eliminate these grain size effects in bedload from rivers of the Amazon basin by measuring $^{10}\text{Be}/^9\text{Be}$ ratios after selective chemical extraction of reactive authigenic phases. These phases mainly comprise Fe-Mn(hydr)oxides that are the main carriers of ^9Be and ^{10}Be . We explore the distribution of ^{10}Be and its stable ^9Be counterpart between these phases and the partitioning of Be into remaining silicate fractions. This extraction procedure was carried out on bedload samples comprising the three main tectonic units of the Amazon basin, namely the Andes, the Guyana and Brazilian Shields, and the central Amazonian lowlands. For all samples, extracted ^{10}Be concentrations show a strong decrease with increasing particle size, with up to 20 times more ^{10}Be extracted from the finest analyzed (<30 μm) grain size when compared to the coarsest (90-125 μm) grain size. We attribute this decrease in ^{10}Be concentrations mostly to subsequent dilution by quartz in coarse bedload. However, when normalized to stable ^9Be concentrations that are also extracted from reactive phases, grain size effects are effectively removed

and the resulting “reactive” $^{10}\text{Be}/^9\text{Be}$ ratio is uniform for all particle sizes. The reactive $^{10}\text{Be}/^9\text{Be}$ ratio thus offers important advantages over methods employing single meteoric ^{10}Be concentrations, such that this ratio can now be developed as quantitative tracer for Earth surface processes.

1.) Introduction

Cosmogenic $^{10}\text{Beryllium}$ (^{10}Be) is a rare, radioactive isotope whose presence on Earth is exclusively due to the interaction of high-energy cosmic radiation with target nuclei in the atmosphere (“meteoric” production) and on Earth’s surface in mineral lattices (“*in situ*” production). Its half-life of 1.39 Myr (Chmeleff et al., 2010; Korschinek et al., 2010) makes ^{10}Be well-suited for many Earth surface applications. In the past decade, research has focused on *in situ* ^{10}Be , as concentrations of this variety of the nuclide routinely disclose ages and rates of landforms and processes at the Earth surface (summarized by e.g. Gosse and Phillips, 2001; Bierman and Nichols, 2004; von Blanckenburg, 2005; Granger and Riebe, 2007). The production rate of *in situ*-produced ^{10}Be within the surface of the lithosphere is, however, orders of magnitudes lower than the production of meteoric ^{10}Be in the atmosphere. Recently, the use of meteoric ^{10}Be was revived (summarized by Willenbring and von Blanckenburg, 2010a). The advantages of using meteoric ^{10}Be are, amongst others, the need of much smaller sample amounts, the possibility of application to settings where quartz is absent, and the possibility to measure meteoric ^{10}Be in a large variety of materials, ranging from soil (Pavich et al., 1984; Graly et al., 2010) over lake sediment (Horiuchi et al., 2003), river and marine terraces (Pavich et al., 1986; Maejima et al., 2005), to ocean sediment (Bourlès et al., 1989; Frank et al., 1997; Christl et al., 2003). Another important application is the use of meteoric ^{10}Be adsorbed on sediment as an erosion rate meter in large watersheds. Pioneered by Brown et al. (1988) and You et al. (1988), concentrations of meteoric ^{10}Be measured in sediment at the outlet of a catchment should, similar to *in situ*-produced ^{10}Be , be representative of the denudation rate of the catchment. However, these advantages are compromised by some major, yet largely unexplored, challenges: first, the atmospheric ^{10}Be deposition

rate must be known on a basin-wide scale. For large watersheds, the compilation of ^{10}Be deposition rates of Willenbring and von Blanckenburg (2010a) may be used, which is based on the models by Field et al. (2006) and Heikkilä (2007). Second, the pH-dependent partitioning of Be between solids and water results in the low retention of Be at pH <6 (You et al., 1989; Aldahan et al., 1999). As a consequence of this retention behavior, ^{10}Be concentrations adsorbed to particles are a function of their grain size, as a substantial increase in meteoric ^{10}Be concentration with decreasing sediment grain size has been observed (i.e. in part from using ^7Be , the short-lived isotope, as a proxy for ^{10}Be ; Gu et al., 1996; Wallbrink and Murray, 1996; Blake et al., 2002; Shen et al., 2004). This increase in nuclide concentration has been attributed to the increase in specific surface area and related increase in sorption capacity with decreasing grain size (summarized in Willenbring and von Blanckenburg, 2010a) and to grain size-specific mineralogical composition that governs the presence of adsorption sites (Aldahan et al., 1999). Since finer-grained sediment is usually enriched in clays and other pedogenic minerals, the observed decrease in ^{10}Be concentration may be due to a “dilution” by quartz in coarser-grained sediment.

One way to circumvent the unpredictable retention behavior of ^{10}Be and dilution effects is to normalize its concentration to that of another isotope or element. Attempts have been made using $^{10}\text{Be}/\text{Fe}$ ratios. Helz and Valette-Silver (1992) noted that ^{10}Be concentrations in marine sediments varied by two orders of magnitude, but $^{10}\text{Be}/\text{Fe}$ ratios from the same samples covered a range as low as the analytical error. An obvious choice as normalizing counterpart is the stable isotope ^9Be , since its chemical properties are identical to those of radioactive ^{10}Be . ^9Be , present in crustal rocks in ~ 2 ppm concentrations, is released upon weathering, and mixes with ^{10}Be in the weathering zone (von Blanckenburg et al., 2011). In rivers, the dissolved and the reactive varieties of Be hence form characteristic $^{10}\text{Be}/^9\text{Be}$ ratios (Brown et al., 1992). In lacustrine sediments, the radioactive decay of the $^{10}\text{Be}/^9\text{Be}$ ratio has been used to date sedimentation ages (Lebatard et al., 2010). In the oceans, these isotope ratios when measured in the authigenic phase of marine sediment serve to determine sedimentation rates (Bourlès et al., 1989), reconstruct variations in the Earth's magnetic field (Carcaillet et al., 2004; Ménabéaz et al., 2011), and record the rates of global denudation over the past 10 Myr (Willenbring and von Blanckenburg, 2010b).

In this study, we provide a methodological framework to explore the potential of $^{10}\text{Be}/^9\text{Be}$ ratios in river sediment as a prerequisite to determine basin-wide denudation rates from this ratio (von Blanckenburg et al., 2011). A condition is that grain-size effects affecting the ^{10}Be single isotope system are removed with a reproducible normalization scheme. To this end, we explore a sequential chemical extraction for river sediment that selectively decomposes authigenic phases such as Fe-Mn(hydr)oxides. These phases, precipitated from the water column or from soil solution, carry most of the ^{10}Be (Bourlès et al., 1989) along with a major fraction of ^9Be . We employ this scheme to silt-sized bed sediment from active channels in the Amazon basin, comprising rivers carrying high sediment loads from erosion of the Andes (Beni, Amazon trunk stream), and the sediment-starved rivers draining the slow-eroding cratonic Brazilian and Guyana shields. We characterize our sediment mineralogically by X-ray diffraction, and chemically by X-ray fluorescence analysis. We explore the nature of our chemical extractions by analyzing its composition for major element concentrations with ICP-OES. Altogether, our analyses show that our bedload samples are representative end members of a sediment continuum carried in rivers, the reactive phase is accessible during chemical extraction and mostly comprises Fe-Mn(hydr)oxides, and that the $^{10}\text{Be}/^9\text{Be}$ ratio derived from these phases does not depend on the analyzed grain size fraction.

2.) Methodology

2.1 Samples and study area. Five bedload samples, sampled from the active river bed, were chosen as representative of the three main tectonic units present in the Amazon basin, which are the Andes, the Shields, and the lowlands (see Figure 1 and Tables 1 and 2 for sample localities). The chemical composition of river sediment in the Amazon basin and their rates of production in the Andes and in the cratonic shields have recently been reported by Bouchez et al. 2011 and Wittmann et al. 2011 (for details on samples and sampling procedures see Wittmann et al., 2009 and Wittmann et al., 2011). These samples include: (1) The Beni River, which is a major Amazon tributary in terms of sediment

discharge (mean annual water discharge $Q = 2050 \text{ m}^3/\text{s}$, Guyot et al., 1993) draining the precipitation-rich Bolivian Andes. Thin soils cover the mostly silicic sedimentary rocks in the upper Beni basin, with a clay mineralogy dominated by illite and chlorite (Guyot et al., 2007). Two samples of the Beni close to the Andean piedmont (Be 1 and Be 2) are used here. Values of pH for Beni river water range from 6.1 to 7.8 (Maurice et al., 1999). Sample “Gr 19” from the Grande River represents the dry Bolivian Andes to the south of the Beni basin. For details on sampling procedures and locations for these samples, we refer to Wittmann et al. (2009), who have used the sandy fractions of these samples for *in-situ* ^{10}Be analysis, to derive basin-wide denudation rates of $1340 \text{ t/km}^2/\text{yr}$ for samples Be 1 and Be 2, and $1690 \text{ t/km}^2/\text{yr}$ for Gr 19, respectively.

(2) The slowly eroding cratonic areas of the Guyana Shield and the Brazilian Shield are represented by samples Br 7 and Cb 2, respectively. Both regions are characterized by highly oxidized and weathered soils enriched in iron and aluminum (Allard et al., 2002). Abundances of kaolinite are higher than in most other soils of the Amazon basin, and trace amounts of illite and smectite were measured (Konhauser et al., 1994). These mostly podzolic shield soils mainly consist of sand-sized quartz (Chauvel et al., 1987; Johnsson et al., 1991). Due to tectonic quiescence, the erosion-weathering regime is supply-limited (Johnsson et al., 1991). In general, rivers draining the shields are sediment-starved, hence they contribute less than 10% of the total sediment load carried by the Amazon River (Filizola and Guyot, 2009). The Guaporé River, represented by sample “Cb 2”, features pH-values of 5.3 to 7.3 (Silva et al., 2006) and is one of the largest rivers that drains the Brazilian Shield ($Q = 915 \text{ m}^3/\text{s}$, Coe et al., 2002), joining the central Amazon system via the Madeira River. From the eastern Guyana Shield, sample Br 7 was selected (for sampling coordinates see Table 2) from the Branco River (pH = 6.4 to 7; Allard et al., 2002; Moreira-Turcq et al., 2003; $Q = 2870 \text{ m}^3/\text{s}$, Coe et al., 2002). For details on the latter two samples, we refer to Wittmann et al. (2011), who have used the sandy fractions of these samples for *in situ* ^{10}Be analysis, from which denudation rates of $74 \text{ t/km}^2/\text{yr}$ for sample Cb 2, and $37 \text{ t/km}^2/\text{yr}$ for the upper Branco River (sample Br 7), were, respectively, derived.

(3) Sample “Obi”, taken at Óbidos, is representative of the central Amazon lowland system. At the Óbidos gauging station, the Amazon basin covers an area of more than $6 \times 10^6 \text{ km}^2$, thus being the

world's largest river basin in terms of water discharge (Q of 209,000 m³/s, Latrubesse et al., 2005), and being one of the largest in terms of sediment discharge (an average denudation rate from *in situ*-¹⁰Be yielded ca. 650 t/km²/yr by Wittmann et al., 2011). At Óbidos, we determined a pH value of 6.9, which is in agreement with values determined by Gaillardet et al. (1997). Seasonally flooding of “varzea” soils in the central Amazonian lowlands results in nutrient-rich soils, with major elemental concentrations (e.g. Mn, Mg, Ca, K) being closer to upper crustal abundances when compared to the depleted non-flooded Central Amazon or Shield soils (Konhauser et al., 1994; Guyot et al., 2007). Here, an increasing trend in iron- and manganese oxide and kaolinite contents and a decreasing trend in illite content from near the Andes in the east to central Amazonia in the west has been observed (Guyot et al., 2007), marking increasing chemical weathering intensities.

2.2 Samples and treatment, extraction procedure, and isotope and elemental analysis. All samples were manually dry-sieved from a <125 µm fraction into silty grain size fractions (30-40, 40-62, 62-90, and 90-125 µm) and the remaining silt-clay fraction of <30 µm. Dry sieving was applied to avoid chemical alteration of samples. We applied Be-extraction procedures to all samples to separately analyze ¹⁰Be, ⁹Be, and major element concentrations in the extracted and the remaining mineral-bound fractions. We present all concentrations (at/g_{solid} for ¹⁰Be and ng/g_{solid} for ⁹Be) in square brackets, and ratios (based on at/g_{solid}; dimensionless) in round brackets. The extraction of adsorbed, reactive phases (containing what is called in this study as [¹⁰Be]_{reac} and [⁹Be]_{reac}) and the residual, mineral-bound beryllium (called here [⁹Be]_{min}, and to some extent also containing [¹⁰Be]_{min}) applied here is based on the sequential extraction procedure by Tessier et al. (1979). We adopted this procedure to extract Be-bearing phases (that neglects the explicit recovery of CaCO₃-bearing phases) since we expected low CaCO₃ contents for our detrital sediment samples (see Figure 2). In addition, we distinguished between amorphous and crystalline iron-oxy-hydroxide phases (Wiederhold et al., 2007). Accordingly, our extraction method, for which between 0.5 and 0.75 g of dried, sieved sediment were used (see Tables 1,2), involves five steps: 1) Extraction of the exchangeable “Ex” fraction by using 1M magnesium-chloride (MgCl₂) solution, 2) Extraction of the reactive amorphous oxy-hydroxide “Am-Ox” fraction by

using 0.5M HCl (Fantle and DePaolo, 2004; Wiederhold et al., 2007). This treatment would also dissolve the CaCO_3 that is present in the sample, but most likely does not dissolve silicate minerals (Guelke et al., 2010). In case of a reactive response of the sample to HCl, indicating presence of CaCO_3 , again 10 ml of HCl was added to avoid buffering effects, as 10 ml 0.5M HCl is not able to dissolve >0.25 g CaCO_3 . A minor reaction of carbonate after addition of hydrochloride acid was only observed in case of “Be” samples. 3) Extraction of crystalline oxides “X-Ox” by using hydroxylamine-hydrochloride solution (1M $\text{NH}_2\text{OH} \times \text{HCl}$ in 1M HCl; see Wiederhold et al., 2007). 4) Extraction of the organic “Org” phase (containing e.g. organic litter) by treating the remaining solids with a mixture of 0.01M HNO_3 and 10M H_2O_2 . 5) Full quantitative decomposition of the remaining silicate residue “Min” is not dependent on amount or type of chemicals used; here, we mostly used a mixture of 28M HF and 14M HNO_3 , and in some cases, addition of Aqua Regia (mixture of HCl and HNO_3 at a ratio of 2:1) was necessary to fully decompose the sample, and sometimes we applied microwave digestion to decompose particularly insoluble residues.

Between extraction steps, the sample residues were washed 3 to 4 times with “MilliQ”- H_2O . From all leachates and from the final decomposed silicate residue, aliquots were taken before carrier addition, which were set aside for stable ^9Be and other elemental analysis. For further processing, ^{10}Be aliquots were spiked with 100 to 300 μg of our in-house “Phenakite” ^9Be carrier, which was determined to contain a $^{10}\text{Be}/^9\text{Be}$ ratio of $2.51 \pm 0.13 \times 10^{-15}$. From hereon, separation of ^{10}Be was carried out according to the simplified separation scheme of von Blanckenburg et al. (1996 and 2004), by carrying out anion and cation column separation and alkaline precipitation. $^{10}\text{Be}/^9\text{Be}$ ratios were measured at the accelerator mass spectrometer (AMS) of the laboratory of Ion Beam Physics, ETH Zuerich, and corrected as described by Kubik and Christl (2010; see Appendix for more details on the AMS standards used). Blanks were subtracted and their errors were propagated into all concentrations. The calculated ^{10}Be concentrations with combined analytical and blank errors are given in Tables 1 and 2. Stable ^9Be measurements, major element, and minor element measurements were carried out by using an optical emission, inductively-coupled plasma spectrometer (ICP-OES, model Varian 720-ES with axial optics). We validated the accuracy of measurements by running aliquots of the geostandard “granite CRGP GA”

(Centre de Recherches Pétrographiques et Géochimiques, origin Andlau, Bas-Rhin, France, see Govindaraju, 1995). Internal consistency was checked by processing a second batch of samples after 12 months that yielded reproducible ^9Be results. X-ray fluorescence (XRF) and qualitative X-ray diffractometry (XRD) were applied to powdered bulk (i.e. non-leached but sieved) grain size fractions of samples Be 1, Obi, Br 7, and XRF was carried out for these samples and sample Cb 2 (see Appendix for more details on XRF and XRD procedures).

3.) Results and Discussion

3.1 Sequential extraction and comparison to the Amazon data of Brown et al. (1992). Similar to the approach of Bourlès et al. (1989), we first report the distribution of beryllium in the sequentially extracted fractions. This was done by taking the 30-40 μm fraction of samples Be 2, Gr 19, Obi, and Cb 2, for which all extracted fractions and the silicate residue were analyzed for ^{10}Be and ^9Be (see Table 1). Major element data for these samples are given in Table A.1 (Appendix). The exchangeable (Ex) and the organic (Org) leachates do not contain significant concentrations of either ^{10}Be or stable ^9Be (see Figure 3). For samples Be 2, Gr 19, and Obi, $[\text{}^9\text{Be}]_{\text{EX}}$ were below detection limit, and the $[\text{}^{10}\text{Be}]_{\text{EX}}$ for Gr 19 was also below detection limit. Only sample Cb 2 contained significant $[\text{}^{10}\text{Be}]_{\text{EX}}$ with $330 \times 10^4 \text{ at/g}_{(\text{solid})}$. Most ^{10}Be is located in both oxide leachates (sum of Am-Ox and X-Ox, denoted in the following as $[\text{}^{10}\text{Be}]_{\text{reac}}$; see Figure 3A), relative to $[\text{}^{10}\text{Be}]_{\text{total}}$ (denoting all extracted fractions plus silicate residue). $[\text{}^{10}\text{Be}]_{\text{total}}$ for all samples ranges from 550×10^4 to $14,600 \times 10^4 \text{ at/g}_{(\text{solid})}$ (see Table 1), and $[\text{}^9\text{Be}]_{\text{total}}$ ranges from 930 to 1,440 $\text{ng/g}_{(\text{solid})}$ (see Tables 1,2). 47 to 74% of $[\text{}^{10}\text{Be}]_{\text{total}}$ is located in the amorphous oxide fraction (Am-Ox), and 20 to 45% is located in the crystalline oxide fraction (X-Ox) (see Table 1). The silicate residue contains most of the ^9Be , and $[\text{}^{10}\text{Be}]_{\text{min}}$ is below $100 \times 10^4 \text{ at/g}_{(\text{solid})}$ (see Table 1). Our $[\text{}^9\text{Be}]_{\text{total}}$ data (average ca. 1,200 ng/g , where 64-74% of the total ^9Be is located in the silicate residue, see Figure 3B) are lower than that measured by Brown et al. (1992), who measured concentrations of 3,400 to 4,000 ng/g (see their Table 2). Upper continental crust values range from 1,300 to 3,100 ng/g (Taylor and McLennan, 1995; Rudnick and Gao, 2004), and thus our samples are

somewhat depleted in ^9Be compared to UCC or Brown et al.'s data. We attribute this depletion to the quartz-rich nature of our bed load samples (see Sections 3.5 and 3.6 for discussion), which do not contain ^9Be -enriched clay minerals like smectites (Grew, 2002).

Brown et al. (1992) obtained similar ^{10}Be distributions among the leachates, and also similar concentration ranges, with ^{10}Be in their hydroxylamine-leached phase ("HLPP") ranging between 1,000 and $23,000 \times 10^4$ at/g_(solid) (n = 7). Our ^{10}Be _{reac} ranges between 300 and $31,000 \times 10^4$ at/g_(solid) for all samples (see Tables 1,2A). Comparing ^{10}Be _{min} of Brown et al.'s (1992) study and ours also yields similar results: we obtain ^{10}Be _{min} of between 30 and $2,300 \times 10^4$ at/g_(solid) (see Tables 1,2B), and Brown et al. (1992) obtained ^{10}Be _{min} of 400 - $1,600 \times 10^4$ at/g_(solid) (n = 3). Brown et al. (1992) hypothesized that a significant portion of this ^{10}Be _{min} may be *in situ*-produced. However, since detrital (although coarser-grained) Amazonian quartz contains *in situ* ^{10}Be concentrations (^{10}Be _{insitu}) of 4.3 - 47×10^4 at/g_(Qtz) only (Wittmann et al., 2011), this ^{10}Be _{insitu} can be regarded as negligible relative to meteoric ^{10}Be concentrations (^{10}Be _{met}). It seems more likely that this ^{10}Be _{min} is due to ^{10}Be hosted in pedogenic clays that were not dissolved during either Am-Ox or X-Ox steps of the sequential extraction (Barg et al., 1997).

3.2 Grain size test. Our results from the sequential extraction procedure (Section 3.1) show that ^{10}Be is primarily located in the two oxy-hydroxide (Am-Ox and X-Ox) extracts. Next we explore how these ^{10}Be fractions are distributed between different grain sizes. To this end, we combined Am-Ox and X-Ox leachates, which were separately extracted, and, after aliquots for major element and ^9Be analysis were taken, combined and measured as one ^{10}Be sample (denoted as " Be_{reac} "). These combined oxide extractions and the remaining silicate residue of Be 1, Obi, Br 7, and Cb 2 were analyzed on the <30, 30-40, 40-62, 62-90, 90-125, and 125-250 μm grain sizes (the latter only for sample Obi; see Table 2).

^{10}Be _{reac} is clearly dependent on particle size (see Figure 4A). Sample Cb 2 shows the most pronounced decrease in nuclide concentration with increasing grain size; sample Be 1 shows the lowest variability in

meteoric ^{10}Be concentrations. The slight increase in $[\text{}^{10}\text{Be}]_{\text{reac}}$ for Obi's 125-250 μm grain size fraction is regarded as an outlier. The silicate residue of most samples does not contain significant amounts of ^{10}Be (see Figures 3 and 4B). One exception is sample Br 7, in which $[\text{}^{10}\text{Be}]_{\text{min}}$ is about equal to $[\text{}^{10}\text{Be}]_{\text{reac}}$ (see Fig. 4B), so that the sum of both oxide leachates and residue is about a factor of 2 higher than $[\text{}^{10}\text{Be}]_{\text{reac}}$ alone.

$[\text{}^9\text{Be}]_{\text{reac}}$, and, to a lesser extent, $[\text{}^9\text{Be}]_{\text{min}}$ show a pronounced decrease with increasing grain size (Fig. 4C,D). For all grain sizes, the distribution of ^9Be among the oxide extracts and the silicate residue agrees with our extraction results for the 30-40 μm grain size fraction (see Section 3.1), with most of the total ^9Be located in the silicate residue (see Table 2). The discussion of these results will follow in Section 3.4.

3.3 Comparison of the chemical composition of bedload and suspended load. In this section, we explore whether our set of samples, which represent coarse sediment sampled on the river bed, are representative proxies of bulk river sediment forming a grain size continuum with finer suspended sediment that was differentiated by sorting. If true, then our samples are part of this chemical and mineralogical continuum that spans the whole range of sediment grain sizes transported by the studied rivers.

To do so, our grain size-specific bedload data are compared to an extensive dataset on bulk bedload and suspended particulate matter (collected at various depths in the river channel) of the Amazon River at Óbidos and the Beni River at its Andean outlet. We focus here on $[\text{}^9\text{Be}]_{\text{total}}$ and $[\text{Fe}]_{\text{total}}$ (taken from Bouchez et al., 2011; see Table A.3), because Fe is the main constituent of Fe-(hydr)oxides that make up the reactive phase. Both were plotted as a function of the Al/Si ratio (Figure 5). This ratio is a chemical proxy serving as a substitute for particle size: the lower the ratio the coarser the particles (Galy et al., 2008; Bouchez et al., 2011). We use this proxy for grain size in absence of other grain size indicators (such as the median D90, which can be used if samples are not, as in our case, pre-sieved). Most bedload data plots at Al/Si ratios <0.2 , which is in accordance with (1) XRD data showing high abundance of quartz in our samples and (2) their coarse grain size typical for river bed sediment (see Section 3.4

and Bouchez et al., 2011). Suspended load is characterized by higher Al/Si ratios >0.2 , evidently being enriched in Al from clay minerals. Both Be and Fe concentrations increase with increasing Al/Si, forming a single trend, albeit with significant variability, in which sieved bedload samples overlap with non-sieved bulk and suspended sediment data. This seamless transition from bed- to suspended load suggests that river sediment can be described by a binary mixture between a coarse and a fine end member, characterized by enrichment of quartz and clay minerals, respectively. The continuum in the distribution of $[\text{Be}]_{\text{total}}$ and $[\text{Fe}]_{\text{total}}$ (and also e.g. Na, K, not shown here) with grain size is visible in the Amazon system, and to some extent also in the Beni system. In the Beni River, $[\text{Fe}]_{\text{total}}$ of our sieved bed sediment samples is somewhat higher than those of bulk bedload and suspended sediment samples, for a given Al/Si ratio. This difference could be, for example, due to temporal changes in sediment source area within the Andean hinterland of the upper Beni basin (Wittmann et al., 2009), as sieved bedload samples were sampled in October 2002, and bulk bedload and suspended sediment were sampled in April 2007. Altogether, this analysis shows that regarding Be, silt-sized sieved bed sediment samples are representative of the whole range of erosion products transported by the Amazon stream.

3.4 The chemical and mineralogical composition of reactive and bulk phases. Next we explore the chemical and mineralogical nature of the reactive phase retrieved by our leaching procedure, which should not attack phases other than mainly Fe-Mn(hydr)oxides (Tessier et al., 1979). Determining the absolute chemical composition of reactive oxy-hydroxides is impossible as their mass is not determined during the leaching procedure. Exploring the chemical composition of reactive phases is thus done by normalizing reactive element concentrations to the same element in bulk sediment (see Fig. 6). The chemical and mineralogical characterization of bulk sediment phases was carried out by XRF (see Table 3) and XRD (see Table 4).

In Figure 6, we present for each grain size and each river the fraction of Be and major elements (Al, Fe, Ti, Mn, Na, K, Ca) carried by the reactive phase ($[\text{Element X}]_{\text{reac}}$, from combined Am-Ox and X-Ox steps, see Table A.1) relative to elemental concentrations derived from bulk samples ($[\text{Element X}]_{\text{total}}$, i.e. sieved, but not leached). From this assessment (Fig. 6), we can deduce that the reactive amounts of Fe,

Mn, and Be usually comprise the largest fraction of the total sample with the exception of the Branco River (Fig. 6C). Proportions of Na, Ti and K in the reactive phase are low (usually between 2 and 12%, except in Br 7), and intermediate for Ca and Al (between 10 and 30%, with exception of the Beni, see below). Importantly, these observations indicate at first order that the extraction method applied selectively decomposes Be-bearing, reactive Fe-Mn(hydr)oxides in Amazon, Beni and Guaporé river sediment, and that silicate minerals are, to a large extent, not affected by this extraction. The presence of e.g. Al in the leached phases might result from dissolution of gibbsite, as gibbsite is a common mineral in the Amazon River and its tributaries (Allard et al., 2002). Reactive Ca can be a significant component of total Ca especially in Beni river sediment (see Section 2.2), which is consistent with the fact that this river drains in part carbonate lithologies; however, the presence of gibbsite and carbonate could not be verified by XRD analyses (see Table 4). In some of the samples small but detectable amounts of Ca, Na and K in the reactive phase could also be interpreted by low-degree leaching of feldspars and hornblende, as these minerals were detected in our samples by XRD analyses (see Table 4). Detection limits by XRD are however potentially impaired by high quartz abundances, which may prevent clay and other minerals (gibbsite, carbonate) to be detected.

A different pattern is observed for the Branco River (Figure 6C). The $[\text{Si}]_{\text{total}}$ of 40 to 45% in bulk Branco sediment samples (Table 3) and the $[\text{Ti}]_{\text{total}}$ of 0.5 to 3% show that this sample consists mainly of quartz (note that quartz contains ca. 46% Si) and Ti-bearing minerals such as illmenite, titanite and rutile. XRD results indeed show the presence of these minerals in the bulk fraction of this sample. The overall low reactive fractions of most metals (including those of ^9Be compared to other samples) could thus be due to the lack of Fe-Mn(hydr)oxides, as it would be the case in highly-weathered, quartz-enriched Shield soils where many metals have been lost into solution or have been complexed by organic acids. Alternatively, however, the very high contents of accessory minerals suggest that this sample is unique in terms of its mineralogy, and potentially might simply represent selectively entrained rather than well-mixed bedload (Slingerland, 1977).

3.5 Potential causes for variations of reactive and total Be concentrations with grain size. The concentration of Be (and other metals) in the reactive and the bulk phases might change with grain size for two reasons: (1) dilution by quartz, which is more abundant in larger grain size fractions. Dilution will simultaneously change $[^{10}\text{Be}]_{\text{reac}}$, $[^9\text{Be}]_{\text{reac}}$, $[^9\text{Be}]_{\text{min}}$, and $[^9\text{Be}]_{\text{total}}$. (2) Grain size-dependent changes in the chemical and mineralogical composition of the reactive phase itself. This effect would affect $[^{10}\text{Be}]_{\text{reac}}$, $[^9\text{Be}]_{\text{reac}}$ and $[^9\text{Be}]_{\text{total}}$ (but not $[^9\text{Be}]_{\text{min}}$). Since both these effects change $[^9\text{Be}]_{\text{reac}}$ and $[^9\text{Be}]_{\text{total}}$, the $(^9\text{Be}_{\text{reac}}/^9\text{Be}_{\text{total}})$ ratios presented in Fig. 6 are potentially controlled by both effects, which we aim at disentangling in the following. The observed increase of $[^9\text{Be}]_{\text{total}}$ (sum of $[^9\text{Be}]_{\text{min}}$ and $[^9\text{Be}]_{\text{reac}}$) with increasing Al/Si ratio (Section 3.3, Fig. 5) might result, at least partially, from dilution by Be-free quartz (Bouchez et al., 2011), which would then also affect $[^9\text{Be}]_{\text{reac}}$ (see discussion below). Indeed, in all rivers, reactive elemental concentrations (see Table A.2) are lower for coarse grain sizes than for finer grain sizes, suggesting that reactive phases are less abundant in coarser-grained sediment. Regarding the effect on $[^9\text{Be}]_{\text{total}}$ by quartz dilution, we can correct this by normalizing $[^9\text{Be}]_{\text{total}}$ to another element not contained in quartz. In Figure 7A, $[^9\text{Be}]_{\text{total}}$ in each grain size fraction is shown normalized to $[\text{Fe}]_{\text{total}}$. The variability of $(^9\text{Be}/\text{Fe})_{\text{total}}$ (and also that of $(^9\text{Be}/\text{Al})_{\text{total}}$, not shown) is much lower than that of $[^9\text{Be}]_{\text{total}}$ alone (Fig. 5A), indeed indicating that $[^9\text{Be}]_{\text{total}}$ is mostly controlled by the dilution of Be-free minerals like quartz and feldspar. This observation agrees with the results of our XRD analysis (Table 4), where in the Amazon and Beni rivers, the coarser fractions all show increasing quartz contents and other minerals such as chlorite, muscovite, orthoclase, albite, and hornblende are less abundant in coarse grain sizes than in smaller grain size fractions.

Regarding the composition of the reactive phase, we proceed similarly by normalizing $[^9\text{Be}]_{\text{reac}}$ to e.g. $[\text{Fe}]_{\text{reac}}$. The relatively uniform $(^9\text{Be}/\text{Fe})_{\text{reac}}$ ratios (Fig. 7B) observed for all grain size fractions of a given sample suggest only slight changes in chemical composition of the reactive phase itself (process 2) with sediment grain size. This furthermore shows that process (1)- dilution by quartz- is the most important factor in determining $[^9\text{Be}]_{\text{total}}$ and hence $[^9\text{Be}]_{\text{reac}}$. By comparing $[^9\text{Be}]_{\text{reac}}$ to $[^9\text{Be}]_{\text{min}}$ (the latter is not contained in quartz), we can ultimately test for the effect of dilution by quartz on $[^9\text{Be}]_{\text{reac}}$. $(^9\text{Be}_{\text{reac}}/^9\text{Be}_{\text{min}})$ ratios vary within a factor of 2-3 for each sample (Fig. 8) except in the Branco. This

variability cannot be due to quartz dilution, but might rather depend on the release rate of $^9\text{Be}_{\text{reac}}$ from Be-containing minerals during weathering. For the Branco, results so far indicate very low and also highly variable $[\text{}^9\text{Be}]_{\text{reac}}$ when compared to $[\text{}^9\text{Be}]_{\text{total}}$ (Fig. 6) and also non-uniform $(^9\text{Be}_{\text{reac}}/^9\text{Be}_{\text{min}})$ ratios (Fig. 8). Regarding the sample's peculiar mineralogical composition, however, these trends might be the result of ^9Be enriched in the clay-free coarse-grained silicate residue (leading to $[\text{}^9\text{Be}]_{\text{min}}$ and $[\text{}^9\text{Be}]_{\text{total}}$ that are “too high” for given $[\text{}^9\text{Be}]_{\text{reac}}$). This can be the case for example if Be-containing minerals are hydrodynamically enriched in the river bed at the sampling location.

This assessment shows that overall, grain size-dependent chemical variability of the reactive phase (process (2)) is probably of minor importance as compared to dilution by quartz (process (1)) for the Beni, Guaporé and Amazon River samples.

3.6 Eliminating grain size effects of ^{10}Be concentrations. The best candidates to correct for grain size-dependent effects on $[\text{}^{10}\text{Be}]_{\text{reac}}$ are concentrations of any element contained in the reactive phase, provided that the change in the composition of the reactive phase with grain size does not become too significant. If such composition effects are minor, any $(^{10}\text{Be}/\text{Element X})_{\text{reac}}$ ratio will remove quartz dilution effects. As shown in Figure 7C-D using the $(^{10}\text{Be}/\text{Fe})_{\text{reac}}$ and the $(^{10}\text{Be}/\text{Al})_{\text{reac}}$ ratio removes most of the grain size effect observed in Figure 4, although some variability remains in the $(^{10}\text{Be}/\text{Fe})_{\text{reac}}$ ratio. Most uniform results are so far obtained with Al_{reac} (Fig. 7D). The remaining variability, which displays only little systematics, is probably due to slight changes in the composition of the reactive phase, as observed in Section 3.5 for $[\text{}^9\text{Be}]_{\text{reac}}$.

Obviously the stable isotope ^9Be is the best candidate for normalization. Note that since the partition coefficient between water and the reactive phase is identical for two isotopes of the same element (ignoring the effects of mass-dependent isotope fractionation), the partitioning of Be between the reactive and the dissolved phase is canceled out by using $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$.

Regarding the observed grain size-dependency, this normalization also cancels out effects derived from both changing the amount of the reactive phase in the sediment (process 1 in Section 3.5) and also changing the composition of the reactive phase (process 2). A quantification of the contribution of each

of these two effects would be desirable, but tedious due to their potential interdependency. We suggest a test to be carried out on artificial sediment as mono-mineralological as possible, in order to eliminate its influence on Be distributions among different particle size fractions. In such a test, the effect of particle size on Be adsorption by the increase in specific surface area as opposed to additional effects of the specific mineralogic composition (Aldahan et al., 1999) could be evaluated. So far, Be adsorption tests have been performed on natural sediment and loess, without paying special attention to the mineralogical composition of the samples (Gu et al., 1996; Shen et al., 2004).

The normalization used for ($^{10}\text{Be}/^9\text{Be}$) is removing the decrease of $[^{10}\text{Be}]_{\text{reac}}$ with increasing grain size (Fig. 4), and resulting ratios are uniform throughout all grain sizes and all samples, even for sample Br 7 despite its peculiar mineralogy. The standard deviation calculated after averaging ratios over all grain size fractions is within the range of associated individual analytical uncertainties (Table 2).

4.) Conclusion

In this study, we present the operational approach to derive ($^{10}\text{Be}/^9\text{Be}$)_{reac} ratios by sequential chemical extractions from silty-sandy Amazonian river bed sediment. We explore the distribution of ^{10}Be and ^9Be among the extracted phases (which are exchangeable, amorphous oxide, crystalline oxide, organic, and silicate residue phase), and find most meteoric ^{10}Be (90-97% of the total ^{10}Be) to be located in both cumulated oxide phases, which extract the surface-reactive amorphous and crystalline Fe-Mn-(hydr)oxides. The silicate residue contains the most stable ^9Be . From bulk analysis, we find that our samples represent the typical bed load chemistry of the major Amazonian watersheds, with cations (e.g. Fe and Be) showing a linear increase in concentration with increasing Al/Si ratio, a proxy for grain size. These grain size effects can be mostly explained by an enrichment of quartz with increasing grain size, while effects of grain size-dependent changes in chemical composition of the reactive phases are probably subordinate. For cumulated reactive oxide leachates, extracted ^{10}Be concentrations also show a strong decrease with increasing particle size. We explore means to normalize these effects. Reactive Fe and Al yield a satisfactory normalization, despite widely differing degrees of weathering of the four

explored Amazonian river systems. When normalized to reactive ^9Be concentrations, the grain size effects on ^{10}Be concentrations are most effectively removed. This normalization results in $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ ratios that are uniform between all grain sizes for a given river sediment sample. Provided that we improve our understanding of the pathways of meteoric ^{10}Be and stable ^9Be during weathering and erosion, this system provides the means to obtain grain size independent rates of Earth surface processes in the coming future.

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Figure captions

Figure 1: Location of samples in the Amazon basin, denoting the main tectonic units and major tributaries.

Figure 2: Sequential extraction procedure for separating reactive from silicate-bound beryllium. The full extraction procedure can be found in the Appendix A.2.

Figure 3: Results of sequential extractions of the 30-40 μm grain size fraction for ^{10}Be (A, $\text{at/g}_{\text{solid}}$) and ^9Be concentration measurements (B, $\text{ng/g}_{\text{solid}}$), calculated relative to the initial solid sample weight (see Table 1). Extracted fractions are Ex (exchangeable), Am-Ox (amorphous oxides), X-Ox (crystalline oxides), Org (organic phase), and Min (silicate residue). At the bottom of each plot, the relative ranges

of ^{10}Be and ^9Be fractions, respectively (in percent), are given. For Ex-extraction step, three samples showed ^9Be concentrations below detection limit (DL). All uncertainties are 1σ .

Figure 4: A) $[^{10}\text{Be}]_{\text{reac}}$ (at/g_{solid}; from combined Am-Ox and X-Ox extractions) for different grain size fractions (from <30 to 250 μm) relative to the initial solid sample weight (see Table 2). B) $[^{10}\text{Be}]_{\text{min}}$ (at/g_{solid}). Significant amounts of ^{10}Be are usually not contained in the silicate residue. C) Stable $[^9\text{Be}]_{\text{reac}}$ (ng/g_{solid}) from combined oxide extracts and D) $[^9\text{Be}]_{\text{min}}$ (silicate residue) for different grain size fractions (from <30 to 250 μm , see Table 2). In all plots, Br 7 30-62 μm (see Table 2) was plotted instead of “30-40 μm ” in absence of this grain size fraction. Wherever possible (see Table 2), we calculated ^9Be concentrations from the average of batch 1 and 2. 1σ uncertainties are mostly smaller than the symbol size.

Figure 5: A) $[^9\text{Be}]_{\text{total}}$ concentrations of sieved bulk samples for Beni, Amazon, Branco and Guaporé Rivers (denotes the sum of $[^9\text{Be}]_{\text{reac}} + [^9\text{Be}]_{\text{min}}$) and non-sieved bedload and suspended load (“SPM”) data (see Table A.3 for summary on dataset) for the Amazon River at Óbidos and the Beni River. B) Total $[\text{Fe}]_{\text{total}}$ for this study’s sieved bulk sediment, in comparison with Fe data from non-sieved bedload and suspended sediment from Bouchez et al. (2011) for the same rivers. UCC denotes upper continental crust values of Rudnick and Gao (2004). Individual sample uncertainties are not shown, but the method-specific external reproducibility is around 10%.

Figure 6: Fractions (%) of reactive elemental concentrations from combined oxide leaching steps ($[\text{Element}]_{\text{reac}}$; Fe, Mn, Be, Al, Ca, Na, Ti, and K, measured by ICP-OES) relative to bulk XRF-derived (“total”) concentrations of the same element, both calculated relative to initial sample solid weight. Average values (%) for each element (gray bar) are also given. $[\text{Na}]_{\text{total}}$ and $[\text{Be}]_{\text{total}}$ were calculated using the sums of “reac” and “min” in the absence of XRF-derived data. The 30-40 μm grain size fractions of all samples as well as the Cb 2 <30 μm fraction could not be analyzed by XRF due to limited sample amounts; thus the former are only given for Na and Be. Results for Mg were not included, as the “Ex” step involved the addition of MgCl_2 , which potentially carried over into the following extraction steps

that are presented here. For sample Be <30 μm , the $[\text{Ca}_{\text{reac}}]/[\text{Ca}_{\text{total}}]$ resulted in a value >100%, which we attribute to inter-method discrepancies. Sorting of elements on the x-axis follows the decrease of reactive fractions for the Amazon River at Óbidos. Individual sample uncertainties are not shown, but the method-specific external reproducibility is around 10%.

Figure 7: Ratios of ($^9/^{10}\text{Be}/\text{Element}_{\text{reac} / \text{total}}$) derived from normalization of $[\text{Be}]_{\text{reac}}$ and $[\text{Be}]_{\text{total}}$ (A, B; both in $\text{at/g}_{\text{solid}}$), and $[\text{Be}]_{\text{reac}}$ (C, D; in $\text{at/g}_{\text{solid}}$) to total and reactive Fe and Al concentrations for different grain size fractions (from <30 to 250 μm). $[\text{Fe}]_{\text{total}}$ is XRF-derived. Note that XRF data is not available for all samples; and no data was obtained for the reactive fraction of Obi <30 μm . 1σ uncertainties are mostly smaller than the symbol size. Concentration results of sample Obi 125-250 μm are regarded as outliers. For this sample, heterogeneity of the aliquots taken for XRF and sequential extraction, respectively, probably causes observed trends.

Figure 8: Ratio of ($^9\text{Be}_{\text{reac}}/^9\text{Be}_{\text{min}}$) derived from $[\text{Be}]_{\text{reac}}$ ($\text{at/g}_{\text{solid}}$) normalized to $[\text{Be}]_{\text{min}}$ ($\text{at/g}_{\text{solid}}$). For most samples, relatively uniform ratios evolve. 1σ uncertainties are mostly smaller than the symbol size.

Figure 9: (A) ($^{10}\text{Be}/^9\text{Be}_{\text{reac}}$) from normalization of $[\text{Be}]_{\text{reac}}$ to $[\text{Be}]_{\text{reac}}$ concentrations for different grain size fractions (from <30 to 250 μm). 1σ uncertainties are mostly smaller than the symbol size. (B) Same dataset normalized to the finest sample (“<30 μm ”). Uncertainties of samples >30 μm are summed from individual 1σ errors and corresponding error of the finest sample.

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Table 1: ^{10}Be and ^9Be data for sequentially extracted fractions

Sample	Lat/Long (° UTM)	"ZB" AMS code (for ^{10}Be results)	Grain size fraction (μm)	Extracted fraction	Initial solid sample weight (g)	^{10}Be concentration ^a ($\times 10^4$ at/g _{solid})	^9Be concentration ^b (ng/g _{solid})
Be 2	-14.2844 / -67.4737	ZB7989	30-40	EX	1.2518	2.0 $\pm$ 2.9	<i>B.D.</i>
OBI	-1.9359 / -55.4989	ZB7987	30-40	EX	1.2626	5.0 $\pm$ 4.9	<i>B.D.</i>
Gr 19	-18.9091 / -63.4095	ZB7988	30-40	EX	1.2125	<i>B.D.</i>	<i>B.D.</i>
Cb 2	-13.4829 / -61.0446	ZB7986	30-40	EX	1.2031	327 $\pm$ 21	3.90 $\pm$ 0.23
Be 2		ZB7863	30-40	AM-OX	1.2518	277 $\pm$ 22	193.3 $\pm$ 9.7
OBI		ZB7861	30-40	AM-OX	1.2626	1456 $\pm$ 60	240 $\pm$ 12
Gr 19		ZB7862	30-40	AM-OX	1.2125	299 $\pm$ 18	287 $\pm$ 14
Cb 2		ZB7860	30-40	AM-OX	1.2031	9971 $\pm$ 299	248 $\pm$ 12
Be 2		ZB7867	30-40	X-OX	1.2518	264 $\pm$ 21	180.2 $\pm$ 9.0
OBI		ZB7865	30-40	X-OX	1.2626	395 $\pm$ 32	119.9 $\pm$ 6.0
Gr 19		ZB7866	30-40	X-OX	1.2125	194 $\pm$ 16	236 $\pm$ 12
Cb 2		ZB7864	30-40	X-OX	1.2031	4225 $\pm$ 165	101.6 $\pm$ 5.1
Be 2		ZB7871	30-40	ORG	1.2518	3.8 $\pm$ 3.8	0.17 $\pm$ 0.10
OBI		ZB7869	30-40	ORG	1.2626	0.75 $\pm$ 3.5	0.60 $\pm$ 0.11
Gr 19		ZB7870	30-40	ORG	1.2125	11.6 $\pm$ 3.6	3.55 $\pm$ 0.20
Cb 2		ZB7868	30-40	ORG	1.2031	83 $\pm$ 19	0.86 $\pm$ 0.11
Be 2		ZB7437	30-40	MIN ^c	0.5055 [0.5885]	45 $\pm$ 16	[947 $\pm$ 46]
OBI		ZB7441	30-40	MIN ^c	1.2626 [0.6105]	100 $\pm$ 22	[979 $\pm$ 48]
Gr 19		ZB7932	30-40	MIN ^c	1.2125	42.1 $\pm$ 7.1	910 $\pm$ 45
Cb 2		-	30-40	MIN ^c	[0.5708]	- -	[573 $\pm$ 29]
Total Be concentrations (sums of all extractions and silicate residual)						^{10}Be concentration ^a ($\times 10^4$ at/g _{solid})	^9Be concentration ^b (ng/g _{solid})
Be 2						591 $\pm$ 67	1321 $\pm$ 65
OBI						1957 $\pm$ 123	1340 $\pm$ 66
Gr 19						546 $\pm$ 47	1436 $\pm$ 72
Cb 2						14600 $\pm$ 505	928 $\pm$ 46

Note: *B.D.* = Below detection limit; "EX"- Exchangeable fraction; "AM-OX" Amorphous oxide fraction; "X-OX" Crystalline oxide fraction; "ORG" Organic fraction; "MIN" Silicate residue fraction.

^a Calculated from AMS ratio using the sum of ^9Be carrier added and natural ^9Be content (with value given in column " ^9Be conc." of this Table) and blank, with combined analytical and blank error (all uncertainties 1σ), relative to initial solid sample weight.

^b External reproducibility at 1σ level denotes a 5% analytical error of OES instrument for Be concentration measurements.

Values given denote mostly batch 1 analyses; except for values in brackets that are batch 2 analyses.

^c Be in silicate residue (except for sample Gr 19) was measured from second batch samples for which sample weight is given in brackets.

Table 2 A: ^{10}Be and ^9Be data for grain size test on combined AM-OX and X-OX ("reac") fractions

Sample	Lat/Long	"ZB" AMS code (for ¹⁰ Be results)	Grain size fraction	Initial solid sample weight	[¹⁰ Be] _{reac} ^a	[⁹ Be] _{reac} ^b	(¹⁰ Be/ ⁹ Be) _{reac} ^c
	(° UTM)		(μm)	(g)	(x10 ⁴ at/g _{solid})	(ng/g _{solid})	(x10 ⁻¹⁰)
Be 1	-14.5273 / -67.4969	ZB7787	<30	0.6060	1370 ± 75	668 ± 28	3.06 ± 0.25
		ZB7429	30-40	0.4987	691 ± 46	415 ± 18	2.49 ± 0.22
		ZB7788	40-62	1.0165	534 ± 26	410 ± 18	1.95 ± 0.15
		ZB7789	62-90	0.9421	626 ± 41	399 ± 17	2.35 ± 0.21
		ZB7790	90-125	1.0267	547 ± 25	353 ± 15	2.32 ± 0.18
average ± std. deviation =							2.43 ± 0.40
OBI	-1.9359 / -55.4989	ZB7791	<30	0.6310	3240 ± 111	644 ± 32 ^d	7.53 ± 0.46 ^d
		**	30-40	1.2626	1851 ± 92	394 ± 17	7.02 ± 0.55
		ZB7792	40-62	0.9510	1080 ± 43	257 ± 13 ^d	6.30 ± 0.40 ^d
		ZB7793	62-90	1.0005	858 ± 38	178.1 ± 7.9	7.21 ± 0.56
		ZB7794	90-125	1.0341	653 ± 35	140.3 ± 6.0	6.96 ± 0.57
		ZB7938	125-250	1.1801	1194 ± 61	239 ± 12 ^d	7.47 ± 0.54 ^d
average ± std. deviation =							7.08 ± 0.45
Br 7	-0.3425 / -61.8022	ZB7795	<30	0.6650	4830 ± 179	279 ± 14 ^d	25.9 ± 1.6 ^d
		ZB7471*	30-62	0.5097	781 ± 56	27.0 ± 1.2	43.4 ± 4.2
		ZB7796	40-62	0.9886	638 ± 40	31.7 ± 1.6 ^d	30.1 ± 2.4 ^d
		ZB7797	62-90	1.0254	432 ± 26	17.77 ± 0.91 ^d	36.4 ± 2.9 ^d
		ZB7798	90-125	1.0191	286 ± 18	15.12 ± 0.79 ^d	28.3 ± 2.3 ^d
average ± std. deviation =							32.8 ± 7.1
Cb 2	-13.4829 / -61.0446	ZB7799	<30	0.5981	30540 ± 917	761 ± 34	60.1 ± 4.2
		**	30-40	1.2031	14200 ± 464	353 ± 16	60.1 ± 4.3
		ZB7800	40-62	1.0032	5630 ± 169	179.9 ± 7.6	46.8 ± 3.1
		ZB7801	62-90	1.0142	2460 ± 136	92.4 ± 3.9	39.8 ± 3.3
		ZB7802	90-125	1.0755	1589 ± 80	42.7 ± 2.1 ^d	55.7 ± 4.0 ^d
average ± std. deviation =							52.5 ± 8.9

Note: This table gives the sums of "Am-Ox" (amorphous oxides) and "X-Ox" (crystalline oxides) fractions (termed "reac")

^a Calculated from AMS ratio using the sum of ^9Be carrier added and natural ^9Be content (with value given in column " $^9\text{Be}_{\text{reac}}$ " of this Table) and blank, with combined analytical and blank error (all uncertainties 1σ), relative to initial solid sample weight.

^b External reproducibility at 1σ level denotes a 5% analytical error of OES instrument for Be concentration measurements; gives average from batch 1 and 2 where possible.

^c Calculated from batch 1 and 2 where possible.

^d Calculated from batch 1 only.

* Denotes grain size of 30-62 μm in absence of 30-40 μm grain size.

** Combined leach was not measured directly but calculated from the sums of individual leaches of Am-Ox (Obi-ZB 7861; Cb 2-ZB 7860) and X-Ox (Obi-ZB 7865; Cb 2-ZB 7864). Corresponding AMS treatment and corrections can be found in Appendix A.1 for each "ZB" number.

Table 2 B: ^{10}Be and ^9Be data for grain size test on silicate residue ("Min")

Sample	Lat/Long (° UTM)	"ZB" AMS code	Grain size fraction (μm)	Initial solid sample weight (g)	$[\text{}^{10}\text{Be}]_{\text{min}}^{\text{a}}$ ($\times 10^4 \text{ at/g}_{\text{solid}}$)		$[\text{}^9\text{Be}]_{\text{min}}^{\text{b,c}}$ ($\text{ng/g}_{\text{solid}}$)		$(^{10}\text{Be}/^9\text{Be})_{\text{total}}^{\text{d}}$ ($\times 10^{-10}$)	
Be 1	-14.5273 / -67.4969	N.A.	<30	[0.5338]	-	-	[1598 $\pm$ 77]		0.90 $\pm$ 0.32	
		ZB7436	30-40	0.4987 [0.5734]	45 $\pm$ 15		[1045 $\pm$ 51]		0.71 $\pm$ 0.26	
		N.A.	40-62	[0.5959]	-	-	[1066 $\pm$ 52]		0.54 $\pm$ 0.19	
		N.A.	62-90	[0.5988]	-	-	[697 $\pm$ 34]		0.85 $\pm$ 0.32	
		N.A.	90-125	[1.0144]	-	-	[581 $\pm$ 28]		0.88 $\pm$ 0.30	
OBI	-1.9359 / -55.4989	ZB7923	<30	0.6310	290 $\pm$ 26		922 $\pm$ 46		3.10 $\pm$ 1.00	
		ZB7441	30-40	0.5020 [0.6105]	100 $\pm$ 22		[979 $\pm$ 48]		2.02 $\pm$ 0.70	
		ZB7924	40-62	0.9510	33.3 $\pm$ 4.7		626 $\pm$ 31		1.83 $\pm$ 0.61	
		N.A.	62-90	[0.6431]	-	-	[592 $\pm$ 29]		1.67 $\pm$ 0.56	
		N.A.	90-125	[1.0535]	-	-	[447 $\pm$ 22]		1.66 $\pm$ 0.58	
		ZB7930	125-250	1.1801	142 $\pm$ 12		548 $\pm$ 27		2.27 $\pm$ 0.79	
Br 7	-0.3425 / -61.8022	ZB7925	<30	0.6650	2300 $\pm$ 113		576 $\pm$ 29		12.5 $\pm$ 4.5	
		ZB7477*	30-62	0.5097 [0.7991]	913 $\pm$ 55		[1727 $\pm$ 85]		1.45 $\pm$ 0.58	
		ZB7926	40-62	0.9886	714 $\pm$ 41		875 $\pm$ 44		2.23 $\pm$ 0.88	
		ZB7927	62-90	1.0254	509 $\pm$ 40		479 $\pm$ 24		2.8 $\pm$ 1.2	
		ZB7928	90-125	1.0191	325 $\pm$ 24		107.2 $\pm$ 5.4		7.5 $\pm$ 3.1	
Cb 2	-13.4829 / -61.0446	N.A.	<30	[0.1330]	-	-	[887 $\pm$ 44]		27.7 $\pm$ 8.8	
		N.A.	30-62	[0.5708]	-	-	[573 $\pm$ 29]		22.9 $\pm$ 7.4	
		N.A.	40-62	[0.6771]	-	-	[525 $\pm$ 26]		12.0 $\pm$ 3.8	
		N.A.	62-90	[0.5130]	-	-	[251 $\pm$ 13]		10.7 $\pm$ 3.8	
		ZB7933	90-125	1.0755	247 $\pm$ 27		97.1 $\pm$ 4.9		17.0 $\pm$ 5.9	

Note: This table gives data for the silicate residue "Min"; N.A. = Not applicable (refers to ZB number for samples that were not measured for ^{10}Be).

^a Calculated from AMS ratio using the sum of ^9Be carrier added and natural ^9Be content (with value given in column " $^9\text{Be}_{\text{min}}$ " of this Table and blank, with combined analytical and blank error (all uncertainties 1σ), relative to initial solid sample weight.

^b External reproducibility at 1σ level denotes a 5% analytical error of OES instrument for Be concentration measurements.

^c ^9Be in silicate residue was measured from batch 1 or 2 samples; the latter values are given in brackets.

^d The $(^{10}\text{Be}/^9\text{Be})_{\text{total}}$ ratio was calculated by $[\text{}^{10}\text{Be}]_{\text{reac}} / ([\text{}^9\text{Be}]_{\text{reac}} + [\text{}^9\text{Be}]_{\text{min}})$, respectively, except for Br 7 samples where "Min" fraction contains significant amounts of ^{10}Be . In that case, we used $([\text{}^{10}\text{Be}]_{\text{reac}} + [\text{}^9\text{Be}]_{\text{min}}) / ([\text{}^9\text{Be}]_{\text{reac}} + [\text{}^9\text{Be}]_{\text{min}})$.

* Denotes grain size of 30-62 μm in absence of 30-40 μm grain size.

Table 3: XRF data for total sediment samples

Sample	Grain size ^a (µm)	Si (%)	Ti (%)	Al (%)	Fe (%)	Mn (%)	Mg (%)	Ca (%)	Na (%)	K (%)	Sr (ppm)	Ba (ppm)	Ni (ppm)	Cr (ppm)	Zr (ppm)
Be 1	<30	35.8	0.90	3.77	4.35	0.07	0.49	0.25	<0.01	1.16	91	581	55	73	2946
Be 1	40-62	34.7	0.94	4.43	4.79	0.07	0.58	0.31	0.1	1.35	106	1006	68	80	6296
Be 1	62-90	35.6	1.04	3.89	5.09	0.08	0.53	0.25	<0.01	1.21	94	695	62	81	2943
Be 1	90-125	39.0	0.72	2.78	3.57	0.06	0.37	0.18	<0.01	0.89	77	244	51	64	620
OBI	<30	27.0	0.70	8.7	5.40	0.09	1.10	1.02	0.76	2.12	220	675	71	108	494
OBI	40-62	34.8	0.76	5.3	3.59	0.06	0.73	1.08	0.72	1.50	198	409	57	86	1235
OBI	62-90	35.4	0.47	5.5	2.76	0.04	0.69	0.99	0.79	1.69	200	511	52	69	170
OBI	90-125	39.3	0.24	3.78	1.50	0.03	0.39	0.63	0.50	1.25	157	337	54	63	72
Br 7	<30	42.8	1.15	0.89	1.06	0.06	0.09	0.06	<0.01	0.44	49	119	36	45	3455
Br 7	30-62	38.7	2.98	1.50	2.48	0.15	0.11	0.07	<0.01	0.84	62	246	37	79	15050
Br 7	62-90	42.8	1.48	0.89	1.28	0.08	0.09	0.05	<0.01	0.48	55	167	43	53	3061
Br 7	90-125	44.5	0.52	0.70	0.48	0.03	0.09	0.04	<0.01	0.39	47	164	35	42	445
Cb 2	40-62	39.7	1.02	2.83	1.24	0.04	0.09	0.13	<0.01	1.03	<10	400	12	57	4637
Cb 2	62-90	43.9	0.46	1.02	0.50	0.02	0.05	0.05	<0.01	0.47	<10	218	<10	26	1154
Cb 2	90-125	45.1	0.11	0.50	0.17	<0.01	0.03	0.02	<0.01	0.22	36	112	12	27	125

^aThe 30-40 µm grain size fractions of all samples and the <30 µm fraction of Cb 2 could not be analyzed for XRF due to very limited sample amounts.

Table 4: Quantitative XRD data for bulk samples

Sample	Grain size (µm)	Quartz content (%)	Other minerals
Be 1	<30	30	Chlorite, Muscovite, Orthoclase, Albite, K-Hornblende
Be 1	40-62	60	same minerals as in Be 1 <30, but less abundant
Be 1	62-90	60	same minerals as in Be 1 <30, but less abundant
Be 1	90-125	75	same minerals as in Be 1 <30, but less abundant
OBI	<30	30	Chlorite, Muscovite, Orthoclase, Albite, K-Hornblende
OBI	40-62	60	same minerals as in OBI <30, but less abundant
OBI	62-90	60	same minerals as in OBI <30, but less abundant
OBI	90-125	75	same minerals as in OBI <30, but less abundant
Br 7	<30	>90	Ilmenite, Titanite, Rutile
Br 7	30-62	>90	same minerals as in Br 7 <30, but less abundant
Br 7	62-90	>90	same minerals as in Br 7 <30, but less abundant
Br 7	90-125	>90	same minerals as in Br 7 <30, but less abundant

Note: Due to limited sample amounts, a qualitative XRD analysis and analysis of Cb 2 could not be carried out.

Appendix

A.1 Details on analytical treatment

Samples with codes ZB7429 to ZB7441 (see Table 1) were measured relative to the standard S555, with a determined $^{10}\text{Be}/^9\text{Be}$ ratio of 95.5×10^{-12} that was compatible with the old ^{10}Be half-life of 1.51 Myr (Hofmann et al., 1987). After April 1st, 2010, the Zuerich AMS lab introduced a new standard, S2007N, with a nominal value of $^{10}\text{Be}/\text{Be} = 28.1 \times 10^{-12}$, that has been calibrated to ICN 01-5-1 (produced by K. Nishiizumi). Samples ZB7787 to ZB7989 were measured relative to S2007N; therefore, we decided to reduce all samples measured before April 1st, 2010, by a factor of 1.096 as suggested by Kubik and Christl (2010) to allow for direct comparison with data obtained relative to the standards produced by K. Nishiizumi et al. (2007). The new standard is also compatible with the newly determined ^{10}Be half-life ($1.387 \pm 0.012 \times 10^6$ yr; Chmeleff et al. 2010; Korschinek et al., 2010).

The external reproducibility of our in-house ICP-OES instrument is within 5% range for all elements (even though the standard deviation of four replicates measured during one analysis of Be was always <0.5% for the most reliable Be wavelength of 313.107 nm, we decided to propagate a 5% error also on Be measurements for external, long-term reproducibility), except for Na and K, where our ICP-OES reproducibility lies within 15% only (see Appendix Tables A.1 and A.2).

Powder X-ray patterns were collected using a Siemens D5000 powder diffractometer with Cu K- α radiation, automatic divergent and antiscatter slits and a secondary graphite monochromator with scintillation counter. The samples were measured from 4° to 75° 2 θ with a step width of 0.02 and a counting time of 4 s per step, with generator settings of 40 kV and 30 mA. The Rietveld algorithm BGMN was used for quantitative analysis (Bergmann et al., 1998). Major element oxides and trace elements were analyzed by XRF on 105 °C-dried samples, prepared as fused disks of Lithium-tetraborate-metaborate (FLUXANA FX-X65, sample-to-flux ratio 1:6). A wavelength-dispersive spectrometer (Panalytical Axios Advanced) and matrix correction programs were used to calculate cation concentrations. H_2O^+ and CO_2 were determined using a high-temperature catalytic combustion analyzing system (Vario EL III).

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A.2 Full extraction methodology

1. Exchangeable Beryllium (Be_{ex}).

- Weigh ca. 1 g of sample.
- Add 10 ml 1M MgCl_2 solution.
- Mild shaking, room temperature, overnight.
- Centrifuge for 15 minutes at 4000 rpm.
- Separate supernate from sediment residue.
- Dry down the supernate at 110°C
- Dissolve in 10 ml of 3M HNO_3 .
- Take aliquots for major and minor element analysis, and for ^{10}Be separation.

2. Amorphous Oxide-bound Beryllium ($\text{Be}_{\text{am-ox}}$).

- Wash sediment remaining from previous step with ultrapure water.
- Add 10 ml 0.5M HCl solution. (Add some more HCl if carbonate minerals are present).
- Agitate gently for 24 hours.
- Centrifuge for 15 minutes at 4000 rpm.
- Separate supernate from sediment residue.
- Dry down the supernate at 110°C close to dryness.
- Dissolve in 10 ml of 3M HNO_3 .
- Take aliquots for major and minor element analysis, and for ^{10}Be separation.

3. Crystalline Oxide-bound Beryllium ($\text{Be}_{\text{x-ox}}$).

- Wash remaining from previous step sediment with ultrapure water.
- Add 10 ml 1M hydroxylamine-hydrochloride solution (in 1M HCl).

- Heat at 80°C for 4 hours with manual overhead shaking every 10 minutes.
- Centrifuge for 15 minutes at 4000 rpm.
- Separate supernate from sediment residue.
- Remove hydroxylamine-hydrochloride solution by adding a mixture of concentrated HNO₃ and H₂O₂.
- Dry down at 70°C. Repeat adding the mixture if necessary until solution is clear.
- Dissolve in 10 ml of 3M HNO₃.
- Take aliquots for major and minor element analysis, and for ¹⁰Be separation.

4. Organically-bound Beryllium (Be_{org}).

- Wash remaining from previous step sediment with ultrapure water.
- Add 2 ml 0.01M HNO₃ and 5 ml 10M H₂O₂ to beaker.
- Place on hotplate at 80°C for 2 hours.
- Add 1 ml H₂O₂ and leave on hotplate for another hour.
- Add 2 ml 0.01M HNO₃ and leave on hotplate for another hour.
- Transfer to tube. Centrifuge for 15 minutes at 4000 rpm.
- Separate supernate from sediment residue.
- Evaporate the supernate close to dryness at 90°C.
- Dissolve in 10 ml of 3M HNO₃.
- Take aliquots for major and minor element analysis, and for ¹⁰Be separation.

5. Silicates and Clay Minerals (Be_{min}).

- Wash remaining from previous step sediment with ultrapure water.
- Transfer to microwave vessel, add about 8 ml HNO₃, 5 ml HCl and 5 ml HF; run microwave.
- Dry down close to dryness at 100°C.
- Second microwave run with Aqua Regia to destroy fluorites.
- Dry down slowly at about 70°C.
- Add 1 ml 3M HNO₃ to beaker, put closed on hotplate at 80°C for ~1 hour
- Label & weigh new centrifuge tube.
- Transfer sample into new tube and add about 8 - 9 ml of 3M HNO₃.
- Dissolve in 10 ml of 3M HNO₃.
- Take aliquots for major and minor element analysis, and for ¹⁰Be separation.

Table A1: Elemental data for all extraction steps

Sample	Leached phase	Al concentration ^a (ppm)	Fe concentration (ppm)	K concentration (ppm)	Ca concentration (ppm)	Mg concentration ^b (ppm)	Mn concentration (ppm)	Na concentration (ppm)	Ti concentration (ppm)
Be 2	EX	2.6 ± 10	1.5 ± 10	203 ± 32	435 ± 24	-	2.2 ± 10	23 ± 11	B.D.
OBI	EX	1.5 ± 10	1.3 ± 10	136 ± 23	1196 ± 61	-	25 ± 10	18 ± 10	B.D.
Gr 19	EX	2.1 ± 11	0.8 ± 11	108 ± 19	1118 ± 57	-	B.D.	80 ± 16	B.D.
Cb 2	EX	8.6 ± 11	2.7 ± 11	122 ± 22	280 ± 18	-	28 ± 12	25 ± 12	B.D.
Be 2	AM-OX	1080 ± 55	4070 ± 200	112 ± 20	1217 ± 62	1349 ± 68	147 ± 13	18 ± 11	2.6 ± 10
OBI	AM-OX	1560 ± 79	5840 ± 290	78 ± 16	1170 ± 59	1502 ± 76	169 ± 13	22 ± 11	10 ± 10
Gr 19	AM-OX	924 ± 48	5530 ± 280	100 ± 19	3140 ± 160	2090 ± 105	339 ± 21	22 ± 12	1.6 ± 12
Cb 2	AM-OX	759 ± 39	2390 ± 120	26 ± 11	32 ± 10	431 ± 24	64 ± 11	1.9 ± 10	1.1 ± 10
Be 2	X-OX	4530 ± 230	15200 ± 760	443 ± 67	35 ± 11	1562 ± 79	71 ± 11	57 ± 14	13 ± 11
OBI	X-OX	5470 ± 270	15600 ± 780	283 ± 44	270 ± 17	1880 ± 95	86 ± 11	77 ± 15	154 ± 13
Gr 19	X-OX	3540 ± 180	18200 ± 910	486 ± 74	71 ± 12	1563 ± 79	96 ± 12	46 ± 13	10 ± 11
Cb 2	X-OX	3550 ± 180	5090 ± 260	180 ± 29	27 ± 10	178 ± 13	13 ± 10	11 ± 10	35 ± 10
Be 2	ORG	17 ± 10	52 ± 11	3.3 ± 10	0.7 ± 10	3.4 ± 10	0.2 ± 10	0.5 ± 10	9.1 ± 10
OBI	ORG	57 ± 11	85 ± 12	14 ± 11	22 ± 11	12 ± 11	0.8 ± 11	3.1 ± 11	155 ± 13
Gr 19	ORG	48 ± 11	322 ± 19	12 ± 10	2.8 ± 10	14 ± 10	1.3 ± 10	1.3 ± 10	0.9 ± 10
Cb 2	ORG	B.D.	30 ± 11	2.9 ± 10	1.9 ± 10	2.8 ± 10	0.8 ± 10	0.2 ± 10	47 ± 11
Be 2 ^c	MIN	185600 ± 700	4860 ± 210	10200 ± 1230	249 ± 15	460 ± 21	39 ± 11	6110 ± 850	3650 ± 180
OBI ^c	MIN	46300 ± 2100	16400 ± 780	10500 ± 1270	7600 ± 360	2550 ± 122	330 ± 19	4740 ± 650	8490 ± 420
Gr 19	MIN	2370 ± 120	499 ± 27	11600 ± 1700	12 ± 10	21 ± 10	B.D.	577 ± 87	484 ± 26
Cb 2 ^c	MIN	36400 ± 1820	7670 ± 380	5610 ± 840	1460 ± 75	1078 ± 57	287 ± 24	1470 ± 220	7990 ± 400

Note: All values (except when indicated) denote results of single measurement from first sample batch (n=1); B.D.= Below detection limit.

"EX"- Exchangeable fraction; "AM-OX" Amorphous oxide fraction; "X-OX" Crystalline oxide fraction; "ORG" Organic fraction; "MIN" Silicate residue fraction.

^a External reproducibility at 1σ level denotes a 5% analytical error of OES instrument for major elements, except for Potassium and Sodium, where the external reproducibility denotes to 15%. These uncertainties were propagated for each Am-Ox and X-Ox extractions, respectively, along with an uncertainty of 1% for each dilution.

^b Due to using MgCl₂ during "EX" leach, Mg concentrations cannot be determined for "EX".

^c The "min" fraction of these samples denotes results of single measurement from second batch (n=1).

Table A2: Elemental data for grain size test on combined AM-OX and X-OX "reac" extraction steps

Sample	Grain size fraction (μm)	Al concentration ^a (ppm)	Fe concentration (ppm)	K concentration (ppm)	Ca concentration (ppm)	Mg concentration (ppm)	Mn concentration (ppm)	Na concentration (ppm)	Ti concentration (ppm)
Be 1	<30	9340 ± 401	37670 ^c ± 1460	704 ± 84	3420 ± 165	5230 ± 188	530 ± 25	111 ± 17	34.6 ± 8.2
	30-40	5550 ± 253	24670 ^c ± 998	678 ± 89	3190 ± 154	3300 ± 142	305 ± 18	109 ± 15	45.6 ± 6.0
	40-62	4900 ± 209	23240 ^c ± 932	464 ± 56	2100 ± 100	2960 ± 107	269 ± 17	65 ± 12	35.5 ± 8.2
	62-90	4240 ± 185	22660 ^c ± 927	410 ± 53	1728 ± 81	2557 ± 92	275 ± 17	63 ± 12	49.5 ± 8.9
	90-125	3530 ± 157	19020 ^c ± 804	291 ± 39	1270 ± 62	1957 ± 72	214 ± 16	44 ± 10	27.7 ± 7.9
OBI	<30	12020 ^b ± 524	N.D.	N.D.	1410 ^b ± 57	5000 ^b ± 190	465 ^b ± 23	143 ^b ± 23	328 ^b ± 21
	30-40	7470 ± 305	21630 ^c ± 825	786 ± 99	2440 ± 102	3160 ± 120	298 ± 18	270 ^c ± 26	188 ± 13
	40-62	5750 ^b ± 238	14140 ^b ± 582	439 ± 58	1378 ^b ± 54	2800 ^b ± 101	131 ^b ± 16	134 ^b ± 23	208 ± 18
	62-90	3640 ± 148	8107 ± 318	257 ± 33	1307 ± 54	1560 ± 59	95 ± 14	116 ^c ± 13	88.3 ± 9.3
	90-125	3240 ± 138	6664 ± 271	241 ± 32	914 ± 37	1264 ± 51	73 ± 14	122 ± 19	63.9 ^b ± 8.9
	125-250	460 ^b ± 27	889 ^b ± 47	382 ± 51	41 ^b ± 15	137 ^b ± 16	12 ^b ± 15	14 ^b ± 15	16 ^b ± 15
Br 7	<30	8050 ^b ± 306	1934 ^b ± 74	175 ^b ± 26	43 ^b ± 15	951 ^b ± 44	7.5 ^b ± 15	10 ^b ± 15	30 ^b ± 15
	30-40	1321 ± 60	339 ± 19	61 ± 12	192 ± 14	20 ± 12	2.2 ± 10	20.2 ± 6.6	92.0 ± 8.0
	40-62	936 ^b ± 39	229 ^b ± 18	31 ^b ± 16	21 ^b ± 16	510 ^b ± 29	3.5 ^b ± 16	2.1 ^b ± 16	81 ^b ± 16
	62-90	602 ^b ± 28	166 ^b ± 16	23 ^b ± 15	7.0 ^b ± 15	493 ^b ± 28	2.7 ^b ± 15	1.4 ^b ± 15	63 ^b ± 15
	90-125	647 ^b ± 29	149 ^b ± 16	20 ^b ± 16	18 ^b ± 15	495 ^b ± 29	1.6 ^b ± 15	2.9 ^b ± 15	29 ^b ± 15
Cb 2	<30	9120 ± 378	14680 ^c ± 533	459 ± 58	942 ± 42	1028 ± 44	178 ± 16	55 ± 12	68.9 ± 9.5
	30-40	4330 ± 183	7080 ^c ± 262	240 ± 32	372 ± 22	436 ± 22	103 ± 15	38 ± 11	41.2 ± 8.4
	40-62	2066 ± 85	3990 ^c ± 147	151 ± 23	283 ± 18	351 ± 21	53 ± 14	35.9 ± 9.4	37.6 ± 8.4
	62-90	2120 ± 100	6110 ^c ± 285	178 ± 30	304 ± 20	688 ± 36	35 ± 14	61 ± 16	73 ± 10
	90-125	783 ^b ± 36	684 ^b ± 29	52 ^b ± 16	6.9 ^b ± 15	431 ^b ± 25	8.7 ^b ± 15	3.8 ^b ± 15	29 ^b ± 15

Note: This table denotes the results of the first and second sample batch (except when indicated) and thus gives the average of two measurements.

"Am-Ox" Extraction step for amorphous oxides, "X-Ox" extraction step for crystalline oxides; N.D. = Not determined.

^aExternal reproducibility at 1σ level denotes a 5% analytical error of OES instrument for major elements, except for Potassium and Sodium, where the external reproducibility denotes to 15%. These uncertainties were propagated for each Am-Ox and X-Ox extractions, respectively, along with an uncertainty of 1% for each dilution.

^bValues denote single measurement from first sample batch (n=1) only.

^cValues denote single measurement from second sample batch (n=1) only.

Table A.3: Summary of dataset used in Figure 5

Sample	Sampling location	Type	Sampling Depth (m)	D90 ^a	Al/Si	Fe concentration (%) ^{b,c}	Be concentration (ppm) ^d
UCC					0.26	3.92	2.10
Be 1 <30	Beni at Rurrenabaque	bedload	-	-	0.11	4.35	2.27
Be 1 40-62	Beni at Rurrenabaque	bedload	-	-	0.13	4.79	1.48
Be 1 62-90	Beni at Rurrenabaque	bedload	-	-	0.11	5.09	1.10
Be 1 90-125	Beni at Rurrenabaque	bedload	-	-	0.07	3.57	0.93
OBI <30	Amazon at Óbidos	bedload	-	-	0.32	5.40	1.57
OBI 40-62	Amazon at Óbidos	bedload	-	-	0.15	3.59	0.88
OBI 62-90	Amazon at Óbidos	bedload	-	-	0.16	2.76	0.77
OBI 90-125	Amazon at Óbidos	bedload	-	-	0.10	1.50	0.59
Br 7 <30	Branco	bedload	-	-	0.02	1.06	0.85
Br 7 30-62	Branco	bedload	-	-	0.04	2.48	1.75
Br 7 62-90	Branco	bedload	-	-	0.02	1.28	0.50
Br 7 90-125	Branco	bedload	-	-	0.02	0.48	0.12
Cb 2-40-62	Guaporé	bedload	-	-	0.07	1.24	0.70
Cb 2-62-90	Guaporé	bedload	-	-	0.02	0.50	0.34
Cb 2-90-125	Guaporé	bedload	-	-	0.01	0.17	0.14
AM-05-44	Amazon at Óbidos	bedload	-	357	0.08	5.89	0.87
AM-05-46	Amazon at Óbidos	bedload	-	195	0.07	1.58	0.94
AM-06-66	Amazon at Óbidos	bedload	-	71	0.26	3.85	2.00
AM-05-35	Amazon at Óbidos	SPM	58	170	0.20	5.17	2.90
AM-05-36	Amazon at Óbidos	SPM	45	165	0.21	3.33	1.63
AM-05-37	Amazon at Óbidos	SPM	30	147	0.24	3.43	1.73
AM-05-38	Amazon at Óbidos	SPM	15	55	0.35	3.88	1.87
AM-05-39	Amazon at Óbidos	SPM	2	82	0.28	4.93	2.40
AM-05-40	Amazon at Óbidos	SPM	43	133	0.23	4.37	2.16
AM-05-41	Amazon at Óbidos	SPM	30	57	0.36	3.71	1.79
AM-05-43	Amazon at Óbidos	SPM	2	61	0.32	5.18	2.59
AM-06-55	Amazon at Óbidos	SPM	55	205	0.21	5.48	3.06
AM-06-56	Amazon at Óbidos	SPM	40	67	0.31	3.48	1.84
AM-06-57	Amazon at Óbidos	SPM	25	56	0.32	4.49	2.54
AM-06-58	Amazon at Óbidos	SPM	10	52	0.33	4.65	2.25
AM-06-59	Amazon at Óbidos	SPM	0	31	0.40	4.66	2.50
AM-06-60	Amazon at Óbidos	SPM	35	66	0.29	5.26	2.84
AM-06-61	Amazon at Óbidos	SPM	20	39	0.35	4.39	2.35
AM-06-62	Amazon at Óbidos	SPM	0	30	0.43	4.82	2.72
AM-06-63	Amazon at Óbidos	SPM	45	58	0.32	5.45	3.25
AM-06-64	Amazon at Óbidos	SPM	20	42	0.36	4.61	2.01
AM-06-65	Amazon at Óbidos	SPM	0	27	0.42	4.93	2.51
BE-07-05	Beni at Rurrenabaque	bedload	-	328	0.10	2.49	1.40
BE-07-10	Beni at Riberalta	bedload	-	175	0.09	2.09	1.27
BE-07-01	Beni at Rurrenabaque	SPM	4.5	213	0.42	5.44	3.09
BE-07-02	Beni at Rurrenabaque	SPM	3	55	0.17	3.36	2.51
BE-07-03	Beni at Rurrenabaque	SPM	1.5	52	0.31	4.62	3.19
BE-07-04	Beni at Rurrenabaque	SPM	0	44	0.32	4.65	3.15
BE-07-06	Beni at Riberalta	SPM	7	109	0.33	4.72	3.33
BE-07-07	Beni at Riberalta	SPM	5	107	0.20	3.66	2.22
BE-07-08	Beni at Riberalta	SPM	2.5	79	0.19	3.42	2.24
BE-07-09	Beni at Riberalta	SPM	0	50	0.23	3.86	2.53

Note: "SPM" denotes suspended particulate matter.

^aD90 gives the prevailing particle size at a cumulative volume percentage of 90%.

^bMeasured using XRF, except samples labelled "AM" and "Be-07", which were measured by ICP-OES.

^cFe data for samples "AM" and "Be-07" are from Bouchez et al., 2011.

^dMeasured using ICP-OES, except samples "AM" and "Be-07", which were measured by ICP-MS.