



Effects of grain size, mineralogy, and acid-extractable grain coatings on the distribution of the fallout radionuclides ^7Be , ^{10}Be , ^{137}Cs , and ^{210}Pb in river sediment

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Abstract

Grain-size dependencies in fallout radionuclide activity have been attributed to either increase in specific surface area in finer grain sizes or differing mineralogical abundances in different grain sizes. Here, we consider a third possibility, that the concentration and composition of grain coatings, where fallout radionuclides reside, controls their activity in fluvial sediment. We evaluated these three possible explanations in two experiments: (1) we examined the effect of sediment grain size, mineralogy, and composition of the acid-extractable materials on the distribution of ^7Be , ^{10}Be , ^{137}Cs , and unsupported ^{210}Pb in detrital sediment samples collected from rivers in China and the United States, and (2) we periodically monitored ^7Be , ^{137}Cs , and ^{210}Pb retention in samples of known composition exposed to natural fallout in Ohio, USA for 294 days.

Acid-extractable materials (made up predominately of Fe, Mn, Al, and Ca from secondary minerals and grain coatings produced during pedogenesis) are positively related to the abundance of fallout radionuclides in our sediment samples. Grain-size dependency of fallout radionuclide concentrations was significant in detrital sediment samples, but not in samples exposed to fallout under controlled conditions. Mineralogy had a large effect on ^7Be and ^{210}Pb retention in samples exposed to fallout, suggesting that sieving sediments to a single grain size or using specific surface area-based correction terms may not completely control for preferential distribution of these nuclides.

We conclude that time-dependent geochemical, pedogenic, and sedimentary processes together result in the observed differences in nuclide distribution between different grain sizes and substrate compositions. These findings likely explain variability of measured nuclide activities in river networks that exceeds the variability introduced by analytical techniques as well as spatial and temporal differences in erosion rates and processes. In short, we suggest that presence and amount of pedogenic grain coatings is more important than either specific surface area or surface charge in setting the distribution of fallout radionuclides.

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1. INTRODUCTION

Tracing and quantifying sediment movement improves the understanding of Earth surface processes (Dietrich et al., 1982) and their interplay with diverse drivers including tectonics (Zeitler et al., 2001), climate (Reiners et al., 2003), and human activity (Hooke, 2000). Quantitative geochemical techniques are increasingly employed for sediment analysis (Guzmán et al., 2013). For example, the fallout radionuclides (FRNs) ^7Be ($t_{1/2} = 53$ d), meteoric ^{10}Be ($^{10}\text{Be}_m$; $t_{1/2} = 1.4$ My), ^{137}Cs ($t_{1/2} = 30.2$ y), and unsupported ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$; $t_{1/2} = 22.3$ y) have been used to estimate erosion rates (Brown et al., 1988; Walling et al., 1999; Matisoff and Whiting, 2012) and to trace sediment sources (Wallbrink and Murray, 1993; Reusser and Bierman, 2010; Belmont et al., 2014; Hancock et al., 2014; Smith and Blake, 2014).

Grain-size dependent variability in FRN concentration has been widely observed in both soils and detrital sediment samples (e.g., He and Walling, 1996; Wittmann et al., 2012; Smith and Blake, 2014; Taylor et al., 2014) and is generally attributed to variations either in specific surface area (He and Walling, 1996; Wittmann et al., 2012; Taylor et al., 2014) or varying mineralogy (Tamura, 1963; Aldahan et al., 1999; Nakao et al., 2014). A third possibility that may also explain observed grain size dependencies is that FRN concentration is a function of grain-coatings (Greene, 2016). Although FRN systems are well-understood in soils (Parsons and Foster, 2011, 2013; Mabit et al., 2013), grain-size dependent variability in nuclide activity of detrital sediments is less frequently studied (Brown et al., 1988; Wallbrink and Murray, 1993; Mabit et al., 2008; Belmont et al., 2014; Hancock et al., 2014).

In this paper, we compare grain size, mineralogy, and composition of acid-extractable materials to the distribution of FRNs (primarily ^{10}Be and ^{210}Pb) in detrital sediment in order to understand better observed grain size dependencies. We do this by measuring nuclide activities in different size fractions of detrital fluvial sediment, and measuring change in nuclide activity over time in a controlled natural fallout experiment with sediment of known grain size and composition. We then quantify the composition of acid-extractable materials as a proxy for the grain coatings in which these nuclides reside using X-ray diffraction (XRD), portable X-ray fluorescence (pXRF), and inductively coupled plasma spectrometry (ICP-OES).

2. BACKGROUND

Fallout radionuclides have different sources. ^7Be and $^{10}\text{Be}_m$ are cosmogenic isotopes created by cosmic-ray induced spallation in the atmosphere (Papastefanou and Ioannidou, 1996). $^{210}\text{Pb}_{\text{ex}}$ is created in the atmosphere by decay of ^{222}Rn (from ^{238}U decay series) that has escaped from Earth materials as a gas (Appleby et al., 1986). ^{137}Cs is an anthropogenic radionuclide. Its presence in sediment is largely the result of atmospheric nuclear weapons testing during the 1960s, with later and less significant releases from accidents at nuclear power plants after

atmospheric testing ceased (Matisoff and Whiting, 2012). After formation or release, FRNs strongly bind to aerosols (Papastefanou and Ioannidou, 1996; Likuku and Branford, 2011; Kristiansen et al., 2012), which are primarily deposited via precipitation in all but the most arid regions, where dry fall dominates (Ioannidou and Papastefanou, 2006). FRNs can be reworked and redeposited as dust by aeolian transport, changing apparent nuclide deposition rates (Ouimet et al., 2015).

Many studies report grain-size dependent distributions of trace metals, including FRNs, with higher concentrations in fine-grained fractions of sediments and soils (e.g., He and Walling, 1996; Wittmann et al., 2012; Smith and Blake, 2014; Taylor et al., 2014), although some studies (e.g., Brown et al., 1988) show little if any grain size effect on FRN concentration in fluvial sediment. Prior research suggests two explanations for preferential FRN distribution that are not mutually exclusive: (1) FRNs are sorbed to particles in proportion to specific surface area, so finer sediment fractions accumulate more nuclides per unit mass (He and Walling, 1996; Wittmann et al., 2012; Taylor et al., 2014); and (2) FRNs are preferentially sorbed to specific minerals (Tamura, 1963; Aldahan et al., 1999; Nakao et al., 2014), and thus finer sediment fractions have higher nuclide activity due to grain-size dependent prevalence of such minerals. Typically, grain size dependencies are accounted for by sieving sediment to a particular grain size (usually <63 μm , though sometimes as small as <10 μm or even <2 μm), limiting analysis to suspended sediment (c. f., Guzmán et al., 2013), or applying a correction factor based on specific surface area (e.g., He and Walling, 1996). The usefulness of specific surface area correction factors is debated (Smith and Blake, 2014). Furthermore, Kaste et al. (2014) found that 100% of ^7Be delivered in rainfall was adsorbed to gravel in a natural delivery experiment, suggesting that coarse grained materials do adsorb some FRNs.

The presence of competing cations may affect the degree of sorption as well (Srivastava et al., 2005). Beryllium may compete with Al^{3+} and Ca^{2+} for sorption sites (Bhat et al., 2005; Graly et al., 2010; Willenbring and von Blanckenburg, 2010). Similarly, Cs competes with K (Steeffel et al., 2003; Fuller et al., 2014) and Pb competes with other heavy metals (Covelo et al., 2007; Seo et al., 2008). Much of the ^7Be and $^{210}\text{Pb}_{\text{ex}}$ in soils and sediments may be bound to, or co-precipitated with, Fe–Mn-(hydr)oxides (Jia et al., 2006). While Wittmann et al. (2012) find that the majority of the $^{10}\text{Be}_m$ inventory in Amazon River bed sediment had co-precipitated with authigenic amorphous and crystalline Fe–Mn-(hydr)oxides, a global meta-analysis of $^{10}\text{Be}_m$ data showed that $^{10}\text{Be}_m$ distribution can be proportional to a variety of soil characteristics but that no single parameter dominates (Graly et al., 2010). Others have found that $^{10}\text{Be}_m$ may eventually become incorporated into authigenic clays (Barg et al., 1997). Fe and Mn phases often form as grain coatings during pedogenesis (Stoops et al., 2010), suggesting that the composition and concentration of grain coatings is likely to be an important factor in FRN retention by sediment. Here, we test that suggestion. Although some have found that organic matter is a

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