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Tracking eolian dust with helium and thorium: Impacts of grain size and provenance

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Abstract

Reconstructions of the deposition rate of windblown mineral dust in ocean sediments offer an important means of tracking past climate changes and of assessing the radiative and biogeochemical impacts of dust in past climates. Dust flux estimates in ocean sediments have commonly been based on the operationally defined lithogenic fraction of sediment samples. More recently, dust fluxes have been estimated from measurements of helium and thorium, as rare isotopes of these elements (He-3 and Th-230) allow estimates of sediment flux, and the dominant isotopes (He-4 and Th-232) are uniquely associated with the lithogenic fraction of marine sediments. In order to improve the fidelity of dust flux reconstructions based on He and Th, we present a survey of He and Th concentrations in sediments from dust source areas in East Asia, Australia and South America. Our data show systematic relationships between He and Th concentrations and grain size, with He concentrations decreasing and Th concentrations increasing with decreasing grain size. We find consistent He and Th concentrations in the fine fraction (<5 μm) of samples from East Asia, Australia and Central South America (Puna-Central West Argentina), with Th concentrations averaging 14 μg/g and He concentrations averaging 2 μcc STP/g. We recommend use of these values for estimating dust fluxes in sediments where dust is dominantly fine-grained, and suggest that previous studies may have systematically overestimated Th-based dust fluxes by 30%. Source areas in Patagonia appear to have lower He and Th contents than other regions, as fine fraction concentrations average 0.8 μcc STP/g and 9 μg/g for ^{4}He and ^{232}Th, respectively. The impact of grain size on lithogenic He and Th concentrations should be taken into account in sediments proximal to dust sources where dust grain size may vary considerably. Our data also have important implications for the hosts of He in long-traveled dust and for the ^{3}He/^{4}He ratio used for terrigenous He in studies of extraterrestrial He in sediments and ice.

We also investigate the use of He/Th ratios as a provenance tracer. Our results suggest differences in fine fraction He/Th ratios between East Asia, Australia, central South America and Patagonia, with ratios showing a positive relationship with

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the geological age of source rocks. He/Th ratios may thus provide useful provenance information, for example allowing separation of Patagonian sources from Puna-Central West Argentina or Australian dust sources. He/Th ratios in open-ocean marine sediments are similar to ratios in the fine fraction of upwind dust source areas. He/Th ratios in mid-latitude South Atlantic sediments suggest that dust in this region primarily derives from the Puna-Central West Argentina region (23–32° S) rather than Patagonia (>38°S). In the equatorial Pacific, He/Th ratios are much lower than in extratropical Pacific sediments or potential source areas measured as a part of this study (East Asia, South America, Australia) for reasons that are at present unclear, complicating their use as provenance tracers in this region.

1. INTRODUCTION

Records of eolian mineral dust flux and provenance preserved in ocean sediments, ice cores and terrestrial deposits offer essential insights into past climates. Past dust fluxes provide information about aridity (Rea, 1994; deMenocal et al., 2000) and sediment supply (Sugden et al., 2009) in dust source areas as well as about the strength of winds responsible for dust entrainment and transport (McGee et al., 2010a). Dust provides limiting micronutrients, especially iron, to the surface ocean and to nutrient-depleted soils (Okin et al., 2004; Jickells et al., 2005); it also impacts the Earth's radiative balance, affecting both surface temperatures and precipitation (Miller et al., 2004). Accurate dust flux reconstructions are required to estimate the importance of these effects as drivers of past climate and biogeochemical changes (e.g., Claquin et al., 2003; Martínez-Garcia et al., 2014).

Reconstructions from terrestrial settings such as loess deposits (e.g., Kohfeld and Harrison, 2003; Sun and An, 2005; Bettis et al., 2003), peat bogs (e.g., Weiss et al., 2002; Kylander et al., 2007; Ferrat et al., 2011; Sharifi et al., 2015) and lakes (e.g., Yancheva et al., 2007; Neff et al., 2008) provide important information about dust emissions and transport close to the source, while dust flux records from polar ice cores offer insights into long-range transport (e.g., Biscaye et al., 1997; Fischer et al., 2007). Records from marine sediments fill the middle ground between these archives, offering broad spatial and temporal coverage as well as direct insights into relationships between dust deposition and marine biogeochemistry. Here we focus on improving dust flux reconstructions from marine sediments, though the data presented have applications in terrestrial sedimentary and ice archives as well.

Dust flux reconstructions from marine sediments require an accurate estimate of the concentration of windblown dust in the sediment. In the past decade, several studies have used the dominant isotopes of helium and thorium, ^{4}He and ^{232}Th, to infer dust concentrations in sediments (e.g., Patterson et al., 1999; Marcantonio et al., 2001a, 2009; Anderson et al., 2006; McGee et al., 2007; Winckler et al., 2008; Serno et al., 2014, 2015). Both isotopes are highly enriched in lithogenic minerals relative to marine biogenic sediments, and they are less susceptible to contamination by volcanic inputs than other markers of lithogenic inputs (e.g., Al, Ti). In sediments in which non-volcanic lithogenic inputs are dominantly eolian, ^{4}He and ^{232}Th can thus provide estimates of eolian dust concentrations. An additional reason for their use is convenience: ^{4}He and ^{232}Th are measured as a part of routine analysis for the minor isotopes ^{3}He and ^{230}Th, respectively, both of which are used for determining accumulation rates in marine sediments (Francois et al., 2004; McGee and Mukhopadhyay, 2013).

In settings in which ^{4}He or ^{232}Th are dominantly derived from eolian dust, both dust concentration and bulk sediment flux can then be derived from a single analysis, allowing calculation of the dust flux (Marcantonio et al., 2001a, 2009; Winckler et al., 2005, 2008; Anderson et al., 2006; McGee et al., 2007; Serno et al., 2014) using the following equation:

$$F_{dust} = \frac{[X]_{sed} \cdot MAR}{[X]_{dust}} \quad (1)$$

where F_{dust} is dust flux, MAR is the mass accumulation rate (or flux) of the sediment (derived from ^{3}He or ^{230}Th data), $[X]_{sed}$ is the ^{4}He or ^{232}Th concentration measured in the sediment and $[X]_{dust}$ is the ^{4}He or ^{232}Th concentration assumed for eolian dust. Converting from ^{4}He and ^{232}Th concentrations to dust concentrations and fluxes thus requires an accurate estimate of ^{4}He and ^{232}Th concentrations in dust. At present, we have a limited understanding of the mean values and variability of these concentrations between source areas or grain size fractions.

Constraints on dust provenance are essential to the interpretation of dust flux records. Knowledge of dust provenance links downwind fluxes to source area conditions and atmospheric circulation patterns. Provenance information also helps constrain dust's past climate impacts, as radiative properties and iron availability differ significantly among dust source areas (Sokolik and Toon, 1999; Dubovik et al., 2002; Schroth et al., 2009). A wide variety of dust provenance tracers exists, including mineralogy (Blank et al., 1985), radiogenic isotopes (Grousset and Biscaye, 2005), trace element ratios (Ferrat et al., 2011; Pourmand et al., 2014), and electron spin resonance signal intensity and crystallinity index of quartz (Sun et al., 2013). Despite this large number of tools, dust provenance in distal regions such as the equatorial Pacific, high-latitude oceans and polar ice sheets remains poorly constrained, in part because of significant overlap in dust source area compositions in common provenance measurements. It has