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Investigating the use of ^{232}Th/^{230}Th as a dust proxy using co-located seawater and sediment samples from the low-latitude North Atlantic

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Abstract

The thorium isotope ratio ^{232}Th/^{230}Th can be measured in seawater and sediment samples, and has been used as a proxy to reconstruct lithogenic fluxes to the oceans for the modern day and the Pleistocene. There has not yet been a systematic study testing the proxy using the ^{232}Th/^{230}Th ratio in seawater and the ratio recorded in the underlying sediment. In this study we use co-located core-top sediments and seawater samples from five seamount sites spanning the tropical North Atlantic to investigate the link between seawater and sediment ^{232}Th/^{230}Th ratios across a range of water depths. Our results indicate that a broad correlation exists between seawater and sedimentary ^{232}Th/^{230}Th ratios. Both seawater and sedimentary ^{232}Th/^{230}Th ratios record a signal consistent with decreasing lithogenic input east to west, from Africa to South America. However, calculated ^{232}Th fluxes for the core-top sediment samples indicate a strong dependence on depth, with up to a factor of ~4 difference from shallow (<600 m) to deep sites (>2900 m). This depth dependence is likely caused by either a deficit of ^{230}Th burial at depth compared to the production in the overlying water column, through addition of ^{232}Th, or by a combination of the two. By comparing seawater and sedimentary ^{232}Th/^{230}Th ratios we derive an apparent fractional solubility of ^{232}Th of 29 ± 3%, in reasonable agreement with the upper end of existing estimates.

Keywords: Thorium; Dust; Sediment; Seawater; Isotope

1. INTRODUCTION

Aeolian dust is an important but poorly quantified part of the climate system. Dust affects the climate through interaction with radiation in the Earth's atmosphere and plays a part in the carbon cycle by delivering key micronutrients, such as iron, to the ocean (Jickells et al., 2005; Maher et al., 2010). Knowledge of present day dust fluxes to the ocean comes from model simulations (Mahowald et al., 2005), direct observations (Lawrence and Neff, 2009) and geochemical data (Measures et al., 2008; Hsieh et al., 2011). Model simulations provide a global picture of dust fluxes to the oceans (Niedermeier et al., 2014), but rely on poorly constrained parameters such as the surface properties of the dust source regions, giving rise to uncertainties in simulated fluxes (Mahowald et al., 2005). Given the relative scarcity of direct observations (Kohfeld and Harrison, 2001), geochemical proxy methods provide a means of testing and improving these models as well as our knowledge of dust fluxes (Anderson et al., 2016).

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A widely used method has been the measurement of Al concentrations in the dissolved phase in seawater e.g. (Measures et al., 2008). This approach draws on the fact that the main supply of Al to the ocean is from continentally-derived lithogenic particles. In remote parts of the ocean, dust is the main source of continental material, and so dissolution of these grains leads to a measurable dissolved Al concentration that is correlated with expected dust input (Hydes, 1983). However, a key difficulty in using dissolved Al to reconstruct dust fluxes is in calculating a local residence time (Hsieh et al., 2011). A related geochemical method that allows for the calculation of a local residence time is the combined measurement of two isotopes of thorium: ^{232}Th and ^{230}Th (Hsieh et al., 2011; Hayes et al., 2013).

Th-232 is a long lived (half-life = 1.4×10^{10} yrs; Holden, 1990) primordial nuclide that is found in the upper continental crust at a concentration of approximately 11 ppm (Taylor and McLennan, 1985). As for Al, ^{232}Th is concentrated in the continental crust, so it can be used to trace lithogenic material with a continental origin. Dissolved ^{232}Th in seawater is therefore assumed to be derived from the dissolution of continental detritus in seawater (Hsieh et al., 2011; Hayes et al., 2013; Deng et al., 2014; Lopez et al., 2015). Given that ^{232}Th provides a means of tracking the input of continental material to the ocean, the only other factor needed in order to calculate a flux of dust is a timescale for ^{232}Th addition. The shorter lived isotope ^{230}Th can be used to provide this timescale. Th-230 has a half life of 75,584 yrs (Cheng et al., 2013), and is sourced from the radioactive decay of ^{234}U in seawater. Th-230 is scavenged rapidly onto sediment particles (although the adsorbed ^{230}Th is thought to maintain a reversible exchange with a pool of dissolved ^{230}Th; Bacon and Anderson, 1982), meaning that ^{230}Th has a short oceanic residence time of up to ~40 yrs (Anderson et al., 1983). The residence time of ^{230}Th in seawater can be derived from measurements of ^{230}Th activity in seawater combined with the known production of ^{230}Th from ^{234}U (Broecker et al., 1973). Making the assumption that ^{230}Th and ^{232}Th share the same residence time, the seawater dissolved ^{232}Th concentration can be combined with the ^{230}Th-derived residence time, to determine a flux of dissolved ^{232}Th. Combining dissolved flux estimates with estimates of the fractional solubility of ^{232}Th from continental material allows a total flux of ^{232}Th to be estimated (Hsieh et al., 2011; Hayes et al., 2013; Deng et al., 2014). Estimates of solubility currently account for a significant source of uncertainty in studies that estimate total ^{232}Th flux from dissolved ^{232}Th fluxes (Hsieh et al., 2011; Hayes et al., 2013; Anderson et al., 2016).

Both these isotopes of thorium have also been measured in sediments in order to estimate lithogenic fluxes in the past (Pourmand et al., 2004; Anderson et al., 2006; McGee et al., 2007; Winckler et al., 2008; Lam et al., 2013; Serno et al., 2014; Costa et al., 2016; Jacobel et al., 2016; Kienast et al., 2016; Williams et al., 2016). In this scenario ^{232}Th measured in sediments is assumed to be sourced only from the input of continental detritus (Pourmand et al., 2004; Anderson et al., 2006; McGee et al., 2007). Far from ocean margins it has been assumed that all ^{232}Th in the sediment (in both adsorbed and lattice-bound pools) is sourced from continental aeolian dust (Anderson et al., 2006; McGee et al., 2007; Hsieh et al., 2011; Hayes et al., 2013; Lopez et al., 2015).

The two thorium based methods described above both make use of the constant input of the isotope ^{230}Th in order to calculate a timescale. Intense scavenging from the water column leads to efficient removal of ^{230}Th. If the flux of ^{230}Th to the sediment is assumed to be equal to the production flux of ^{230}Th in the overlying water column (β, ~0.0267 dpm m^{-3} yr^{-1}; Francois et al., 2004), then the $^{230}Th_{xs}$ activity concentration (in dpm g^{-1}), where ‘xs’ denotes excess ^{230}Th that is not supported by ^{234}U decay in the sediment, is a function of the sediment flux to the seafloor. Therefore measurement of $^{230}Th_{xs}$ in the sediment allows vertical mass fluxes at a given water depth (z, in metres) to be calculated (Eq. (1)). This method of calculating vertical sediment fluxes is termed ‘^{230}Th normalisation’ (Francois et al., 2004). Early studies used this method to calculate lithogenic fluxes in the Atlantic (Francois and Bacon, 1991). By combining the sediment mass flux calculated by ^{230}Th normalisation with the fractional concentration of ^{232}Th in the sediment (in μg g^{-1}), a vertical flux of ^{232}Th (in μg m^{-2} yr^{-1}) to the seafloor can be calculated (Eq. (2)). If the ^{232}Th concentration in lithogenic material is known, then an estimate of the lithogenic flux can be made (e.g. McGee et al., 2007). Recently it has been suggested that a concentration of 14 ppm is appropriate for estimating dust fluxes using this method for most locations receiving fine-grained dust (McGee et al., 2016). If the ^{232}Th/^{230}Th ratio of the adsorbed phase of thorium is known ($(^{232}\mathrm{Th}/^{230}\mathrm{Th}_{xs})_{Ads}$ in μg dpm^{-1}), an adsorbed flux of ^{232}Th can also be calculated (Robinson et al., 2008; Eq. (3)). This ‘adsorbed’ pool of thorium can be accessed by leaching sediments without dissolving the ^{232}Th bearing lithogenic phases (Robinson et al., 2008).

$$\text{Sedimentary mass flux} = \beta * z / ^{230}\mathrm{Th}_{xs} \tag{1}$$

$$^{232}\mathrm{Th\ flux} = \text{Sedimentary mass flux} * {}^{232}\mathrm{Th} \tag{2}$$

$$\text{Adsorbed } ^{232}\mathrm{Th\ flux} = (^{232}\mathrm{Th}/^{230}\mathrm{Th}_{xs})_{Ads} * \beta * z \tag{3}$$

Determination of ^{232}Th and ^{230}Th in ocean sediments and seawater therefore has the potential to provide the basis for estimating fluxes of continental material (both in the past and in modern times). However, there has not yet been a study that combines measurements of ^{232}Th and ^{230}Th in core-top sediments with co-located seawater samples.

In this study, we assess the relationship between modern seawater and core-top sedimentary ^{232}Th/^{230}Th ratios in the tropical North Atlantic Ocean. This area is, in part, beneath the path of dust emitted from the largest dust source on Earth, the Sahara Desert (Goudie and Middleton, 2001; Cakmur et al., 2006). We present measured ^{232}Th/^{230}Th ratios and concentrations in core-top sediments and seawater samples, and compare how the values vary across the basin as well as with water depth. We calculate ^{230}Th-normalised mass fluxes and ^{232}Th fluxes for our sample sites and investigate the spatial and vertical variability of these parameters. We also estimate the