Geochemical fingerprinting of trans-Atlantic African dust based on radiogenic Sr-Nd-Hf isotopes and rare earth element anomalies

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ABSTRACT

Mineral dust is an important component of Earth's climate system and biogeochemical cycles on a global scale. In order to understand the relationship between climate processes in the source areas and the properties of aerosols at distant receptor sites, we must be able to identify the source provenance of dust. Here we present a multiproxy study that characterizes the temporal variability in the geochemical composition of long-range African dust (LRAD) collected between 2003 and 2011 in the trade winds on the Caribbean island of Barbados. We find systematic differences between Sr-Nd-Hf isotopic composition and rare earth element anomalies of individual dust events and evidence of seasonal shifts in dust source activity and transport. These results indicate that coherent geochemical source signatures of LRAD can be preserved even after transport across thousands of kilometers. We investigated the possibility of identifying the potential source areas through comparisons with literature data. However, these data are almost entirely based on measurements of soil and sediment samples; this could lead to biases because of soil-aerosol particle size and composition differences. Nonetheless, our data suggest that many samples are linked to sources in Mali and sub-Saharan regions. Radiogenic Nd-Hf composition of aerosols can potentially be a useful proxy to study the proximity of mineral dust sources to depositional sites. In order to establish firmer links between LRAD and dust source areas, however, we require much more data on the geochemical composition of aerosols from potential source areas in North Africa.

INTRODUCTION

Mineral dust plays an important role in many atmospheric and ocean processes that relate to Earth's climate and biogeochemical processes (Shao et al., 2011). North Africa is the world's largest dust source, emitting an estimated 800 Tg yr^{-1}, ~70% of the global budget (Huneeus et al., 2011). Many studies have shown that African dust is transported in great quantities to the western Atlantic, the Caribbean Basin, the southern United States, and South America after a journey of about a week from Africa (Goudie and Middleton, 2001; Prospero and Lamb, 2003;Evan et al., 2009; Prospero and Mayol-Bracero, 2013). Satellite imagery, back-trajectory analysis, and chemical transport models coupled with ground-based measurements are commonly employed to identify the sources of dust in modern dust plumes (Shao et al., 2011). However, in order to link present-day sources to sedimentary and ice records for paleo-reconstructions, we must characterize dust particles using geochemical and mineralogical proxies (Grousset et al., 1998; Muhs et al., 2007; Trapp et al., 2010; Abouchami et al., 2013; Scheuvens et al., 2013). In a recent compilation of literature data that have been used to associate African dust with sources on the continent, Scheuvens et al. (2013) concluded that a combination of radiogenic Sr-Nd isotopes is best suited for differentiating the sources of aerosols within Africa. To date, nearly all efforts have been focused on measuring geochemical tracers of African dust in proximal and distant marine sediments and a few aerosol samples from dust outbreaks transported across the Atlantic (Rickli et al., 2010; Aarons et al., 2013; Scheuvens et al., 2013; Abouchami et al., 2013; Kumar et al., 2014). In this contribution we present the first record of temporal variability in geochemical composition of long-range African dust (LRAD) using radiogenic Sr-Nd-(Hf) isotopes and rare earth element (REE) anomalies that span the past decade.

MATERIALS AND METHODS

Aerosol samples were collected from 2003 to 2011 at a field station located at Ragged Point, a promontory on the east coast of the island of Barbados (13°09′54″N, 59°25′55″W) (Prospero and Lamb, 2003). The samples were fused with $LiBO_2$ alkali flux to ensure complete dissolution of refractory minerals. A three-stage extraction chromatography scheme was devised to separate Sr, Nd, Hf, and the REEs from interfering matrix elements for high-precision isotope and elemental analysis by multicollector–inductively coupled plasma–mass spectrometry (MC-ICP-MS). The analytical accuracy and precision were assessed by processing certified reference materials. Additional details of the analytical methodology are provided in the GSA Data Repository[1] (see also Pourmand and Dauphas, 2010; Pourmand et al., 2012).

RADIOGENIC Sr-Nd SYSTEMATICS AND REE ANOMALIES

There is a strong seasonality in dust transport from North Africa to Barbados; the greatest transport occurs during boreal summer (Prospero and Lamb, 2003). This seasonality is largely linked to regional changes in dust source activity, associated in part with the latitudinal migration of the West African monsoon and the Intertropical Convergence Zone (Stuut et al., 2005). The shift in source activity is clearly visible in satellite aerosol products (e.g., Adams et al., 2012; Hsu et al., 2012). These changes, coupled with the seasonal migration of the trade wind system, result in a latitudinal shift in the Atlantic aerosol plume, which in summer is centered on the latitude of Barbados, coincident with the peak dust concentrations (Fig. DR1 in the Data Repository). The mass median diameter of Barbados dust particles is ~2–3 μm (Li-Jones and Prospero, 1998) and remains relatively unchanged throughout the year. It is therefore expected that Sr-Nd and REE compositions of dust measured at Barbados primarily vary due to heterogeneity in the aerosols from source regions, not to differences in particle-size sorting during transit (Dasch, 1969; Grousset et al., 1998).

Our first objective was to look for temporal shifts in the geochemical composition of different dust events. Second, we sought to link such changes to changes in sources based on available data from North Africa. Previous attempts using bulk elemental concentrations did not yield substantial differences between dust storms during the summers of 2003–2004 (Trapp et al., 2010). We measured $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, $^{176}Hf/^{177}Hf$, and the REEs in 25 dust events sampled during various seasons from 2003 to 2011 (Tables DR3 and DR4, Fig. DR1). Variations in $^{143}Nd/^{144}Nd$

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[1]GSA Data Repository item 2014250, details of the analytical methodology, data tables, and supplementary figures, is available online at www.geosociety.org/pubs/ft2014.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

and $^{176}Hf/^{177}Hf$ are conventionally reported as ε_{Nd} and ε_{Hf}, which are expressed in deviations from the chondritic uniform reservoir values of 0.512638 ± 0.000005 and 0.282785 ± 0.000011, respectively (Bouvier et al., 2008). The temporal changes in radiogenic isotopes and REE anomalies, summarized in Figure 1, reveal time-resolved variability in radiogenic isotope ratios, model Sm-Nd ages, Eu and Ce anomalies ($Eu/Eu^* = \frac{Eu_N}{\sqrt{(Sm_N \times Gd_N)}}$, $Ce/Ce^* = \frac{Ce_N}{\sqrt{(La_N \times Pr_N)}}$; N, normalized to Post Archean Australian Shale) and La_N/Lu_N ratios. Model Sm-Nd ages represent crustal residence ages of aerosol source rocks based on assuming $^{143}Nd/^{144}Nd = 0.51315$ for depleted mantle and a simple evolution leading to modern $^{147}Sm/^{144}Nd = 0.2137$ (Goldstein et al., 1984; Grousset et al., 1988). Consequently, relative changes in model ages are more meaningful than absolute values.

The ε_{Nd} values and Sm-Nd model ages in Barbados aerosols range from –14.1 to –10.0, and from 1.6 to 2.1 Ga, respectively (Table DR3; Fig. 1). Previous measurements of ε_{Nd} in African aerosols also plot within a similar range (Scheuvens et al., 2013). All samples show similar REE patterns with enrichment in middle REEs and a distinct positive Eu anomaly (Fig. DR5a). The Eu and Ce anomalies vary by 6.0% and 11.4% over the decade of study. The Ce anomalies show a systematic shift to more negative values over the record (Fig. 1). The cause of this trend cannot be identified in the absence of data from potential source areas in North Africa. The ε_{Hf} and La_N/Lu_N also show substantial changes from –11.2 to –6.3, and 33.6%, respectively, throughout the record without noticeable long-term trends. Significant covariation is observed among these proxies; younger Sm-Nd ages correspond with more radiogenic Hf and Nd isotope ratios, less positive Eu anomaly, and higher La_N/Lu_N ratios. With the exception of a sample from September 2009, $^{87}Sr/^{86}Sr$ ratios are less variable in comparison, ranging from 0.71025 to 0.71714 with a mean of 0.7158 ± 0.0025 (2 standard deviations).

The range of variability seen for radiogenic isotopes and REE anomalies is much greater than those expected from analytical uncertainties. This is substantiated by the analysis of three samples collected on three consecutive days during a large dust event in March 2006 (Fig. DR1). With the exception of ε_{Hf}, the variability over the three days is far smaller than that over the entire record. This suggests that the dust sources remained unchanged over this interval, or that aerosols from multiple sources were subsequently mixed during transport. Additional studies are required to characterize the compositional variability (or homogeneity) of individual dust events.

The temporal resolution of our data set also makes it possible to examine changes in the source provenance of dust in North Africa at different times of the year. As shown in Figure 2, with the exception of a single event at the end of April 2010, samples from November–April show distinctly younger Sm-Nd ages, and less positive Eu anomalies compared with events from May–September. To our knowledge, this is the first time the influence of seasonality is revealed in the geochemical composition of LRAD.

It is clear from these results that aerosols from North Africa retain distinct geochemical signatures that could potentially be linked to their sources, even after traveling across the Atlantic Ocean. Transport of LRAD to Barbados, however, can take 5–7 days from the west coast of Africa, and 10 or more days from other potential source areas. In the absence of meteorological data, back-trajectory and satellite observations become increasingly unreliable over this time. In order to use geochemical fingerprinting to reveal the source of particles in modern aerosols and paleodust, we need information on the composition of potential source materials and their geographical distribution. Unfortunately, measurements of radiogenic isotopes in aerosols collected over North Africa are quite rare; and nearly all data are based on bulk samples from surface and subsurface bedrock and sediments (Scheuvens et al., 2013). Differences between particle size distribution of aerosols and soil samples could lead to discrepancies in composition (Mahowald et al., 2013). Furthermore, the precise locations of some of the samples are unknown (Scheuvens et al., 2013). With these limitations in mind, aerosol data of Barbados dust events are within a narrow range of the Sr-Nd isotopic composition compared to the wide distributions measured in soil samples from various potential source areas across North Africa (Fig. 3). Kumar et al. (2014) reported similar Sr-Nd isotopic compositions from aerosol samples collected during the summer of 2008 on Tobago (11°19′40″N, 60°32′30″W) and the U.S. Virgin Islands (17°44′44″N, 64°35′09″W). Similar to the pattern observed in Figure 2, the samples from November–April are distinguished from those collected from May–September and demonstrate an intra-annual shift in Sr-Nd isotope composition. Our best assessment of potential sources of LRAD clusters within Mali and sub-Saharan regions (without Senegal), with possible contributions from Libya and the Bodélé Depression in northern Chad. The Sr isotopes of aerosols from Barbados overlap with the Ca-rich end member from soil samples of the Bodélé Depression while the Nd isotopes do not distinguish between Ca- and Si-rich fractions (Abouchami et al., 2013). The REE patterns of soil samples from the Bodélé also show enrichment in middle REEs (Fig. DR5a); however, their Eu anomalies (Fig. DR5b) do not overlap with aerosol samples from Barbados and dust collected off the coast of West Africa (Muhs et al., 2007; Rickli et al., 2010). Aerosol samples measured over Jordan that were associated with sources in Libya show enrichment in heavy REEs that are not observed in samples from Barbados (Fig. DR5b). A conclusive com-

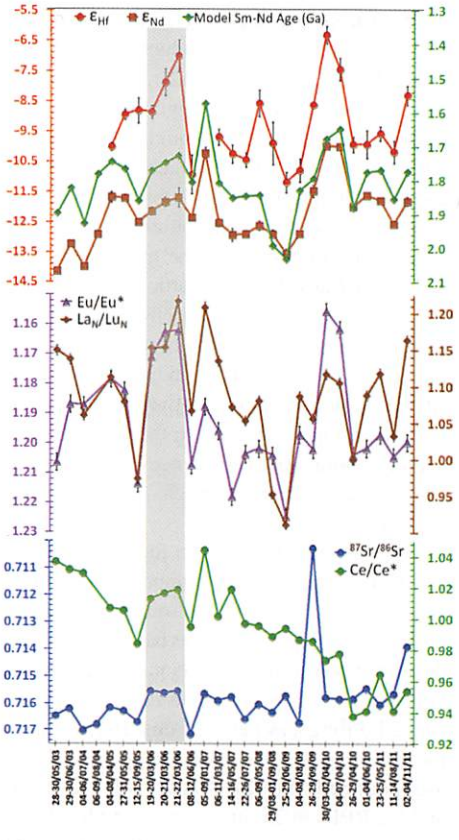

Figure 1. Radiogenic Nd, Hf, Sr isotopes and rare earth element (REE) anomalies show significant variability in trade wind dust samples from Barbados. Data from three consecutive days of March 2006 are shown in gray. Uncertainties on isotope ratios are at 95% confidence interval and relative standard deviation for the REEs. N—normalized to Post Archean Australian Shale.

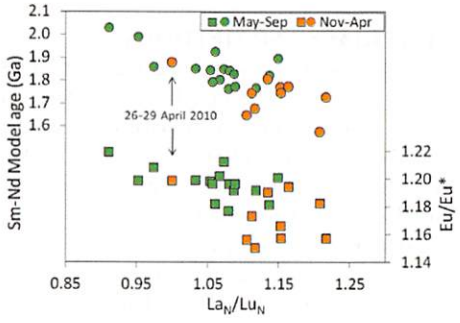

Figure 2. North Africa aerosols collected during May–September in Barbados show marked difference in Sm-Nd model ages and rare earth element anomalies compared with November–April of 2003–2011.

parison cannot be made in the absence of radiogenic isotope and REE data in aerosols from the Bodélé and other regions with vastly different signatures from soils of North African potential source areas (Fig. 3).

RADIOGENIC Hf SYSTEMATIC

The Lu-Hf systematic is complementary to Sm-Nd but $^{176}Hf/^{177}Hf$ ratios have been rarely measured in aerosols and their application to tracking source changes remains largely unexplored. Coupled Nd-Hf isotopes, however, have been used extensively to study continental weathering and modern and paleo-oceanic circulation. The global oceanic budget of Nd and Hf isotopes is a function of riverine and eolian input from the continents, with possible contributions from oceanic crustal sources (Albarède et al., 1998; Vervoort et al., 1999; Lupker et al., 2010). Zircon grains in the coarse sediments have exceptionally high affinity for Hf. This bias leads to low Lu/Hf ratios and a decoupling between radiogenic Nd and Hf isotopes in the world oceans. Consequently, the ε_{Nd}-ε_{Hf} in fine (zircon free) particles measured in ocean sediments are shifted to more radiogenic Hf values relative to crustal and igneous arrays (van de Flierdt et al., 2007; Bayon et al., 2009). Until now, only a few measurements were available, from aerosols over the Atlantic Ocean (Rickli et al., 2010; Aarons et al., 2013) and in Greenland ice (Bayon et al., 2009); nearly all Hf isotope data are from oceanic crust, detrital sediments, and seawater measurements (Vervoort et al., 1999; Bayon et al., 2009; Stichel et al., 2012). The new data from North Africa dust over Barbados presented in Figure 4 fill a large gap in our understanding of the sources of oceanic Hf isotope composition. The aerosols from Barbados align along the zircon-free ε_{Hf}-ε_{Nd} array together with fine riverine sediments and dust particles from the eastern Atlantic Ocean and the Dye-3 ice core (Greenland Ice Sheet Project). Although none of the eolian samples plot on the seawater array, measurements in leachates of Asian desert loess suggest that contributions from the labile fraction of clay-size particles can account for radiogenic Hf composition of seawater without the need to invoke contributions from the oceanic crust (Chen et al., 2013). The influence of zircon grains on Nd-Hf isotope composition of aerosols can potentially be a useful proxy to track changes in wind velocity and/or source proximity to modern and particularly paleodepositional sites. Measurements of Hf isotopes from possible North Africa source areas and along the travel path of aerosols should provide further constrains on the application of this proxy.

CONCLUSIONS

We studied aerosol samples associated with 25 major dust events during the past decade at Barbados and found distinct seasonal shifts and

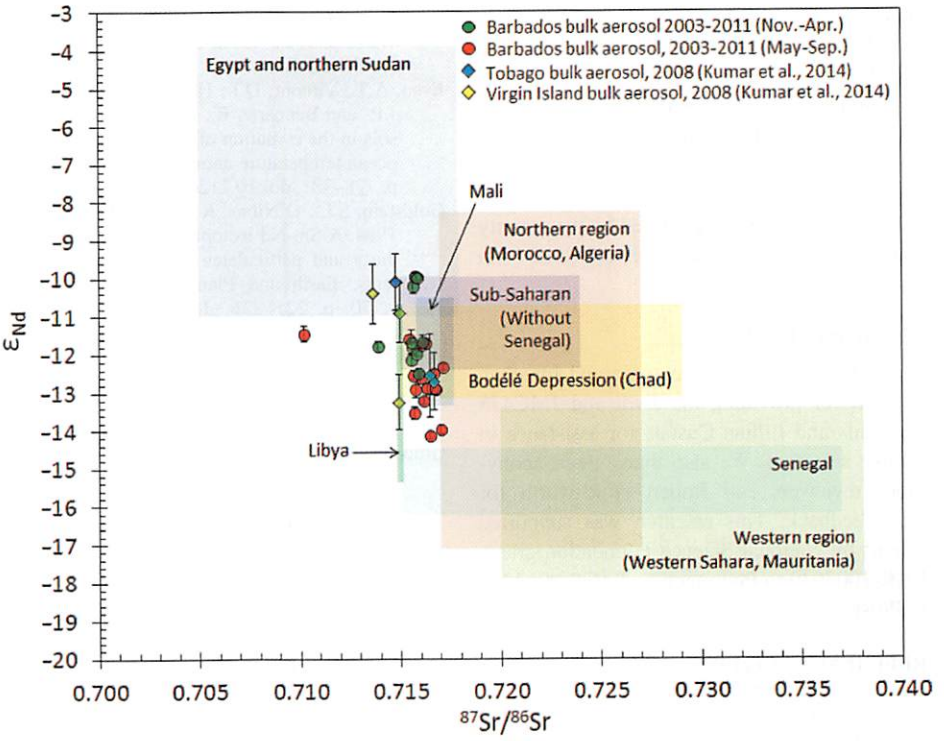

Figure 3. Radiogenic Sr and Nd isotopes in aerosols from Barbados are shown for November–April and May–September. The compositions of summertime aerosols from U.S. Virgin Islands and Tobago are also shown (Kumar et al., 2014). Colored boxes represent ranges of values measured in soil samples of North Africa (Abouchami et al., 2013; Scheuvens et al., 2013). Uncertainties from this study are 95% confidence interval.

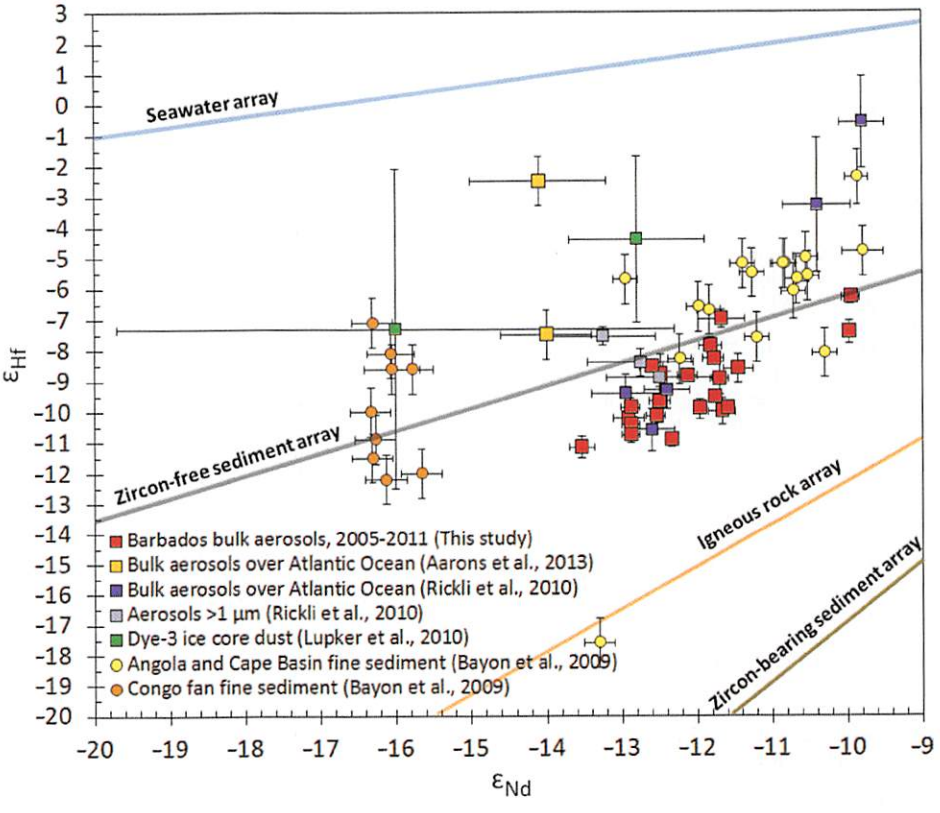

Figure 4. Radiogenic Nd and Hf isotopes in aerosols from Barbados are compared with literature data. The results from Rickli et al. (2010) are based on the mean of multicollector–inductively coupled plasma–mass spectrometry and thermal ionization mass spectrometry replicate measurements. Sediment, seawater, and igneous arrays are reproduced from Bayon et al. (2009). Uncertainties from this study are 95% confidence interval.

significant temporal variability in radiogenic Sr-Nd-Hf isotopes and REE compositions of LRAD. We show that it is possible to tentatively link mineral dust to specific potential source areas in North Africa. However, our ability to use these powerful geochemical tools to identify dust provenances in modern records and paleorecords is currently limited by the scarcity of geochemical data in aerosols collected from potential source regions.

ACKNOWLEDGMENTS

We thank Franco Marcantonio and Jonathan Patchett for providing the JNdi and JMC475 standards and Lillian Custals for assistance in sample selection. We also thank three anonymous reviewers and Robert Holdsworth for their feedback. This research was supported in part by National Science Foundation grants EAR-1003639 to Pourmand and AGS-0962256 to Prospero.

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Manuscript received 28 February 2014
Revised manuscript received 16 May 2014
Manuscript accepted 19 May 2014

Printed in USA