



A radiogenic isotope tracer study of transatlantic dust transport from Africa to the Caribbean



A. Kumar^{a,*}, W. Abouchami^{a,b}, S.J.G. Galer^a, V.H. Garrison^c, E. Williams^d, M.O. Andreae^a

^a Max Planck Institute for Chemistry, P.O. Box 3060, 55020 Mainz, Germany

^b Institut für Mineralogie, WWU Münster, Corrensstraße 24, 48149 Münster, Germany

^c U.S. Geological Survey, St. Petersburg, FL 33701, USA

^d Massachusetts Institute of Technology, Cambridge, MA 02139-4307, USA

HIGHLIGHTS

- Tracing transatlantic dust transport using radiogenic isotopes (Pb, Sr, Nd).
- Natural and anthropogenic sources in African and Caribbean aeolian dust.
- Short-time scale Nd isotopic variability in the aeolian dust.
- Sahel main dust source in spring–summer to the Caribbean.

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ABSTRACT

Many studies have suggested that long-range transport of African desert dusts across the Atlantic Ocean occurs, delivering key nutrients and contributing to fertilization of the Amazon rainforest. Here we utilize radiogenic isotope tracers – Sr, Nd and Pb – to derive the provenance, local or remote, and pathways of dust transport from Africa to the Caribbean. Atmospheric total suspended particulate (TSP) matter was collected in 2008 on quartz fibre filters, from both sides of the Atlantic Ocean at three different locations: in Mali (12.6°N, 8.0°W; 555 m a.s.l.), Tobago (11.3°N, 60.5°W; 329 m a.s.l.) and the U.S. Virgin Islands (17.7°N, 64.6°W; 27 m a.s.l.). Both the labile phase, representative of the anthropogenic signal, and the refractory detrital silicate fraction were analysed. Dust deposits and soils from around the sampling sites were measured as well to assess the potential contribution from local sources to the mineral dust collected. The contribution from anthropogenic sources of Pb was predominant in the labile, leachate phase. The overall similarity in Pb isotope signatures found in the leachates is attributed to a common African source of anthropogenic Pb, with minor inputs from other sources, such as from Central and South America. The Pb, Sr and Nd isotopic compositions in the silicate fraction were found to be systematically more radiogenic than those in the corresponding labile phases. In contrast, Nd and Sr isotopic compositions from Mali, Tobago, and the Virgin Islands are virtually identical in both leachates and residues. Comparison with existing literature data on Saharan and Sahelian sources constrains the origin of summer dust transported to the Caribbean to mainly originate from the Sahel region, with some contribution from northern Saharan sources. The source regions derived from the isotope data are consistent with 7-day back-trajectory analyses, demonstrating the usefulness of radiogenic isotopes in tracing dust provenance and atmospheric transport.

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1. Introduction

It is well established that atmospheric mineral dust is one of the most important constituents in the global aerosol budget and plays

* Corresponding author. Tel.: +49 6131 305 6601; fax: +49 6131 305 6019.

E-mail addresses: ashwini.kumar@mpic.de, ashwinikumarjha@gmail.com (A. Kumar).

a significant role in the overall climate system (Haywood et al., 2003; Usher et al., 2003; Jickells et al., 2005; IPCC, 2007; Ozer et al., 2007; Prenni et al., 2009; Luo et al., 2009; Evan et al., 2011). Based upon estimates reported in Maher et al. (2010) and references therein, more than half of the global dust emission originates from Africa. The Sahara–Sahel dust zone – spanning across ~4000 km from Chad to Mauritania – is the world's largest aeolian dust source (Schütz et al., 1981; d'Almeida, 1989; Swap

et al., 1996). Desert dust provides significant amounts of macro- and micro-nutrients (e.g. phosphate and iron) to the ocean surface, resulting in increased phytoplankton productivity, thereby modulating the carbon cycle of the Earth System (Sarthou et al., 2003; Jickells et al., 2005; Mahowald et al., 2005). African dust has also been suggested to transport pathogens and disease-spreading spores, which can impact the health of coral reefs in the Caribbean (Shinn et al., 2000), as well as semi-volatile organic compounds that may disturb ocean ecosystems (Garrison et al., 2003). Furthermore, African dust is thought to be a significant source of nutrients for the Amazonian rainforest in South America (Reichholf, 1986; Swap et al., 1992; Koren et al., 2006; Bristow et al., 2010; Abouchami et al., 2013).

Using the TOMS (Total Ozone Mapping Spectrometer) absorbing aerosol index, several studies have highlighted the Bodélé Depression, located northeast of Lake Chad, as the most important dust source in the Saharan region (Middleton and Goudie, 2001; Prospero et al., 2002; Koren et al., 2006). The West Sahara region, extending from east of the Mauritanian coast, near the Mali-Mauritanian border to the northeast Mali-Algerian border, is ranked second in dust emission (Engelstaedter et al., 2006). Additional patchy sources of dust emission in northern Africa are in the south of the Atlas Mountains (northern Algeria), the eastern Libyan desert, large areas of Egypt and Sudan and parts of the Sahel (Engelstaedter et al., 2006). Evidence for dust transport from northern Africa towards/across the Atlantic Ocean comes from extensive field campaigns (Prospero, 1999; Reid et al., 2003; Haywood et al., 2008; Ansmann et al., 2011), satellite observations (Koren et al., 2006; Christopher and Jones, 2010; Ridley et al., 2012) and transport models (Ginoux et al., 2004; Schepanski et al., 2009).

The presence of Saharan dust in the Caribbean (Prospero and Lamb, 2003), the south-eastern United States (Perry et al., 1997; Muhs et al., 2007, 2012) as well as South America (Koren et al., 2006; Ben-Ami et al., 2010) reflects long-range transport of dust, which is largely driven by the prevailing atmospheric wind circulation over the African continent. The latitudinal shift of the Inter Tropical Convergence Zone (ITCZ) from summer (located between 15 and 23°N) to winter (located between 5 and 10°N) is a characteristic meteorological feature found over tropical Africa (Engelstaedter et al., 2006). The formation of the Inter Tropical Front (Bou Karam et al., 2008) provides an environment conducive for dust emission, especially over north-western Africa during boreal summer (Engelstaedter and Washington, 2007). However, during boreal winter, the latitudinal shift of the ITCZ allows easterly winds, including the Harmattan, to direct dust-particle transport towards South America (Koren et al., 2006). A combination of low-level jet and Harmattan winds stimulates winter emission of dust from the Bodélé Depression in Central Africa (Washington and Todd, 2005), which has been suggested to be the main source of dust to the Amazon Basin (Koren et al., 2006).

Mineralogical studies of African dust deposits (Caquineau et al., 1998; Stuut et al., 2005; Skonieczny et al., 2011, 2013) have shown that despite being derived from a large geographical source area, African dust has a relatively uniform mineralogical composition (Molinarioli, 1996). Consequently, it is difficult to attribute a characteristic and unambiguous signature to specific source regions. However, naturally-occurring radiogenic isotope systems (e.g. Sr, Nd, and Pb isotopes) are robust tracers of dust sources (Grousset and Biscaye, 2005; Meyer et al., 2011; 2013; Skonieczny et al., 2011; 2013; Abouchami and Zabel, 2003; Abouchami et al., 2013) and can thus provide important information on source provenance and pathways of dust transport. Given the large variability in Sr and Nd isotopic compositions of African source areas (see recent compilation by Scheuven et al., 2013), one would expect to be able

to identify and locate the emission source of dust delivered to the western Atlantic, which is the primary aim of the present study.

We report Sr, Nd and Pb isotopic compositions in mineral dust collected at three locations bordering the Atlantic Ocean in order to identify potential sources and the origin of the dust. Samples were collected in Africa – the putative source region of dust based on earlier descriptions and satellite observations (Prospero et al., 2002) and from sites located on the other side in the Caribbean, where dust transport and delivery typically occur during the boreal summer. The radiogenic isotopic compositions of Sr, Nd and Pb, taken together provide a good characterization of the aeolian dust at the three locations – U.S. Virgin Islands, Tobago and Mali – as well as that of potential local sources. The nature and origin of the dust source(s) are discussed in view of existing literature data on potential African source areas, as well as pathways of dust delivery from Africa to the Caribbean region.

2. Material and methods

2.1. Sampling sites

Total suspended particulate (TSP) samples were collected in spring/summer 2008 on quartz-fibre filters from three different locations (Fig. 1): (A) a dust source region (Sahel): Emetteur Kati, Mali (12.6°N, 8.0°W; 555 m elevation); (B) a downwind site in the Southeast Caribbean: Flagstaff Hill, Tobago (11.3°N, 60.5°W; 329 m elevation); and (C) a downwind site in the Northeast Caribbean: East End, St. Croix, Virgin Islands (17.7°N, 64.6°W; 27 m elevation). During boreal summer, these sampling sites are located within the ITCZ (Fig. 1), which restricts the transport of dust to the southern Atlantic, but the influence of the easterlies allows aeolian transport from the African continent towards the Caribbean. The selected island locations are thus ideal for tracing the provenance of dust during the Northern Hemisphere summer, the isotope signature of which is expected to resemble that of African source(s) in the case of transatlantic transport to the Caribbean.

2.2. TSP sample collection

Quartz microfibre filters (QFFs; numbered 8" × 10" Whatman Grade QM-A Sheets, #1851-865) – preconditioned at 600 °C for 8 h, weighed (Mettler Toledo AB204-S analytical balance), and stored individually in Ziploc bags – were used as the sampling medium at all sites.

In Mali, air was sampled using high-volume brushless electric blower motors (Mercury Northland BBA14 222HMB-00) in custom aluminium tripod enclosures. Each enclosure held two 8" × 10" filter holders with silicon gaskets, a single blower motor and an inline elapsed time and voltage Variac (Anderson Model 09911). On the Virgin Islands and Tobago, air was sampled using a permanent magnetized motor (MAX-SE 3 Phase, TECO-Westinghouse, Round Rock, Texas, USA) to power a HAUCK turbo fan; a computer (ABB, Type ACH401601112) controlled the motor speed and recorded elapsed time and air volume sampled. A Pacific – Sierra Research Corporation air sample holder housed a 60-by-60 cm stainless steel filter holder with silicon gaskets to mask the area to accommodate 6–8" × 10" QFF filters (see above) that rested on a nylon mesh base and overlapped approximately 2 cm. Filters were sealed off from the building housing the equipment; the air intake was approximately 3 m above the building and the exhaust was >2 m downwind of the building at ground level. Samples were collected only when the wind was from NNE to SE in the Virgin Islands and NE to E in Tobago.

Sampling times varied (range 23–63 h, most often 44–48 h) due to logistical constraints, weather (temperatures in Mali), and

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