



# North African mineral dust across the tropical Atlantic Ocean: Insights from dust particle size, radiogenic Sr-Nd-Hf isotopes and rare earth elements (REE)

Michèlle van der Does<sup>a,\*</sup>, Ali Pourmand<sup>b</sup>, Arash Sharifi<sup>b</sup>, Jan-Berend W. Stuut<sup>a</sup>

<sup>a</sup> NIOZ – Royal Netherlands Institute for Sea Research, Department of Ocean Systems, and Utrecht University, Texel, The Netherlands

<sup>b</sup> Neptune Isotope Laboratory, Department of Marine Geosciences, Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, FL, USA

## ARTICLE INFO

### Keywords:

Mineral dust  
Sahara  
Atlantic Ocean  
Sr-Nd-Hf isotopes  
Rare earth elements

## ABSTRACT

Large amounts of mineral dust are exported from North Africa across the Atlantic Ocean, impacting the atmosphere and ocean during transport and after deposition through biogeochemical processes. In order to characterize the isotopic signature of dust from different seasons and years, in relation to their bulk particle size, and to obtain a general idea of its provenance, Saharan dust was collected using subsurface sediment traps moored in the tropical North Atlantic Ocean in 2012–2013, and by shipboard aerosol collection during three trans-Atlantic research cruises in 2005, 2012 and 2015. The samples were analysed for radiogenic Strontium (Sr), Neodymium (Nd), and Hafnium (Hf) isotopes, rare earth element (REE; La-Lu) abundances and particle size. In addition, soil sediments from Mauritania, a potential source area, were analysed and compared to the Atlantic dust samples. The results indicate no relation between Sr and Nd isotopic compositions and dust particle size. In contrast, Hf isotopic compositions show a strong relation with particle size, associated to the so-called zircon effect. We explored alternative sources of lithogenic particles to the sediment traps such as Amazon River sediments. Our results reveal that the sediment-trap samples bear distinctly different geochemical signatures from sediments from the Amazon Basin and Amazon River tributaries, and confirm that the primary source of lithogenic particles is northern Africa. The collected dust samples show close relations to African dust aerosols collected at Barbados and samples from the Bodélé Depression, although differences between seasons are observed, which we relate to differences in source areas.

## 1. Introduction

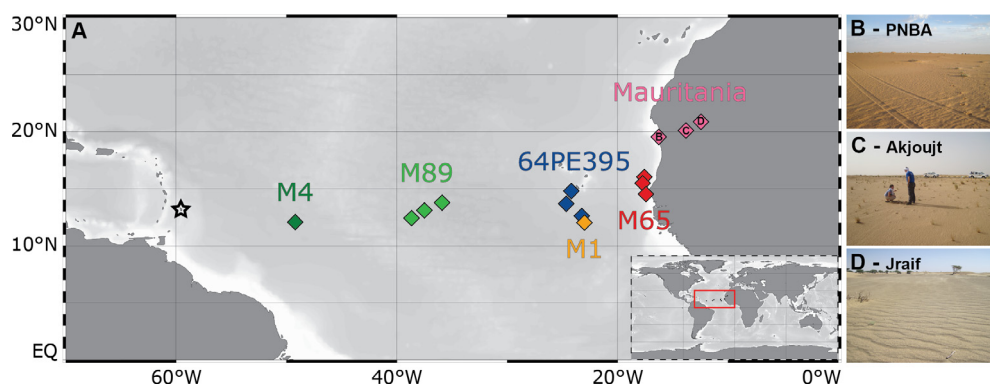
The Sahara Desert in northern Africa is the world's largest dust source (Muhs, 2013), contributing up to 70% of all global annual dust emissions (Maher et al., 2010; Huneeus et al., 2011). In particular, the Bodélé Depression, located in the region of the Lake Chad Basin, is currently the largest single dust source, with the biggest dust export in the world (Koren et al., 2006; Washington et al., 2006), and appears to have been active for at least several hundred to thousands of years (Prospero et al., 2002; Armitage et al., 2015). Nevertheless, the contribution from the Bodélé Depression aerosols transported over the Atlantic basin arriving at Barbados in the Caribbean appears to be small (Pourmand et al., 2014; Bozlaker et al., 2018; Kumar et al., 2018). Most dust emitted from North African sources is transported westward across the Atlantic Ocean, with an estimated amount of 182 Tg between 30°N and 10°S every year (Yu et al., 2015). The emitted dust impacts both the atmosphere and the ocean, from affecting the atmospheric radiation

budget (Ryder et al., 2013) and acting as cloud condensation nuclei and ice nuclei (Wilcox et al., 2010; Atkinson et al., 2013), to the enhancement of the ocean's carbon cycle by delivering nutrients stimulating phytoplankton growth (Martin and Fitzwater, 1988), and by mineral ballasting of organic particles in the ocean (Armstrong et al., 2002; Bressac et al., 2014; Van der Jagt et al., 2018). Saharan dust can also transport viable spores, pathogens and microbes over great distances (Griffin, 2007), which can impact marine and terrestrial ecosystems (De Deckker et al., 2008), and be harmful to human health and increase mortality rates (Morman and Plumlee, 2014; Schweitzer et al., 2018).

Dust particle size and composition vary seasonally, and with the distance over which the dust is transported. Particle sizes decrease downwind as a result of more rapid gravitational settling of coarse-grained particles (Bagnold, 1941; Van der Does et al., 2016), while the mineralogical and isotopic composition of the deposited dust changes downwind by the preferential settling of heavier quartz and zircon particles closer to Africa (Pourmand et al., 2014; Korte et al., 2017). At

\* Corresponding author.

E-mail address: [mdoes@nioz.nl](mailto:mdoes@nioz.nl) (M. van der Does).



**Fig. 1.** A: Location of the samples used in this study in the North Atlantic Ocean: Sediment-traps M1 (12°N, 23°W) and M4 (12°N, 49°W); Soil samples from Mauritania; and average locations of aerosol samples collected during three research cruises: M65, M89 and 64PE395. The Caribbean island Barbados is indicated with a star. Photos of sample locations PNBA (B), Akjoujt (C) and Jraif (D) in Mauritania.

Barbados in the Caribbean, daily dust samples have been collected for over 50 years (Prospero et al., 1970; Prospero and Nees, 1977; Prospero and Lamb, 2003), making it one of the longest present-day dust records. Dust concentrations at this island peak during the summer months, related to the latitudinal movement of the Intertropical Convergence Zone (ITCZ). This leads to a northern displacement of the dust cloud over the Atlantic Ocean in summer, followed by a southward migration in winter (Nicholson, 2000). In addition, mineral dust is typically transported at low altitudes (< 3000 m) by the trade winds during the winter season, whereas the Saharan Air Layer (SAL) carries the dust at higher altitudes (> 5000 m) across the Atlantic during the summer (Stuut et al., 2005; Friese et al., 2016; Van der Does et al., 2016). The latitudinal and altitudinal seasonal shift of the dust-carrying atmospheric systems, in turn, results in a change of dust transport from different source areas and thus a seasonal change of dust composition (Pourmand et al., 2014; Friese et al., 2017).

Compositional characterization of trace elements and different isotope systems of mineral dust particles can yield information about the source area of the dust (Scheuven et al., 2013), and selective processes during transport and deposition such as the sorting based on particle size and shape. Dust provenance identification can be aided by determining the mineralogical composition (Caquineau et al., 1998; Kandler et al., 2009, 2011; Friese et al., 2017), geochemical composition (Moreno et al., 2006; Muhs et al., 2007; Castillo et al., 2008; Bozlaker et al., 2018), identification of biomarkers (Schreuder et al., 2018), and radiogenic isotopes (Grousset and Biscaye, 2005; Meyer et al., 2011; Abouchami et al., 2013; Pourmand et al., 2014). The radiogenic Sr, Nd, Hf isotopes and the REE composition of aerosols at emission and deposition sites are shown to be particularly useful proxies for understanding changes in dust provenances and weathering regimes (Scheuven et al., 2013; Pourmand et al., 2014). Most rare earth elements (REEs) show comparable values relative to the average composition of the upper crust, however the depletion or enrichment of several elements as a result of natural (e.g., chemical and physical weathering processes that lead to enrichment or depletion of heavy minerals such as zircon) or anthropogenic processes can be used as a tracer for source regions (Meyer et al., 2011, 2013; Scheuven et al., 2013).

Previously, Pourmand et al. (2014) analysed Sr-Nd-Hf isotopes and REEs of Saharan dust collected in the Caribbean at Barbados between 2003 and 2011, and demonstrated seasonal shifts of dust sources and transport from the Sahara, as well as a systematic shift in nearly all geochemical proxies as a function of temporally variable dust plumes. Zhao et al. (2018) compared these distal dust samples to surface sediments from the Sahara and Sahel, and also demonstrated the seasonal variation of dust provenance. Here, we present the radiogenic Sr-Nd-Hf isotope and REE composition of Saharan dust collected during three trans-Atlantic research cruises and from submarine sediment traps moored in the tropical North Atlantic Ocean. Their geochemical characteristics will provide a better understanding of dust transported and

deposited over the open Atlantic Ocean. Alternative sources of lithogenic particles to the sediment traps from Amazon River sediments are investigated. By analysing the geochemical composition and combining them with particle-size data, we aim to shed light on the provenance of the lithogenic fraction in the sediment-trap samples of anomalously high depositional events. The data are compared to soil samples collected from Mauritania and to data from literature, to gain insights into the general provenance of the dust, whether that is North African or South American. Moreover, this dataset also allows for comparing the composition of dust particles collected from the atmosphere with those collected after sinking through the water column.

## 2. Material and methods

### 2.1. Sample collection

Saharan dust was collected at five mooring stations along a trans-Atlantic transect, as described by Van der Does et al. (2016) and Korte et al. (2017). The transect consisted of four stations along 12°N, and a fifth station at 13°N, with two sediment traps moored at each station at 1200 m and 3500 m below sea level (BSL). The sediment traps consisted of 24 sampling bottles which were pre-programmed to sample at synchronous intervals of 16 days, from 19 October 2012 to 7 November 2013 (Stuut et al., 2012, 2013). For this study, five samples from three different time intervals were analysed from two sediment traps, moored at 1200 m below sea level at stations M1 (23°W) and M4 (49°W) (Fig. 1, Table 1). These are synchronous sample intervals #12 (13-04-2013–29-04-2013) and #24 (22-10-2013–07-11-2013) from both stations, which showed anomalously high deposition fluxes at M4 (Van der Does et al., 2016), and #9 (24-02-2013–12-03-2013) from M1, which is the sample with the largest dust flux from that station.

Aerosol samples were collected during three different research cruises in the tropical North Atlantic Ocean: FS Meteor Expedition M65 in June 2005 (Mulitza, 2005), FS Meteor Expedition M89 in October 2012 (Stuut et al., 2012), and RV Pelagia Expedition 64PE395 in January 2015 (Stuut et al., 2015) (Fig. 1, Table 1). Samples from these three cruises were picked to represent three different seasons (summer, autumn and winter) of three different years, to shed light on possible differences in provenance of the dust in various years, and to be compared to the dust found in the sediment traps. During all cruises, aerosol sampling was performed with an Anderson high-volume dust collector, mounted on the deck above the bridge of the ship. Dust was collected on letter-sized Whatman Type 41 filters under a protective cover. The samplers were equipped with a wind vane which switched off the sampler during winds blowing at an angle > 90° from the ship's heading. This was done to prevent contamination from the ship's exhaust behind the sampler (see e.g. Stuut et al., 2012).

Soil samples were collected at various sites in Mauritania, one potential source and transit area of mineral dust, during a field campaign in 2009. These are named PNBA, Akjoujt and Jraif (Fig. 1), representing

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