



## Seasonal radiogenic isotopic variability of the African dust outflow to the tropical Atlantic Ocean and across to the Caribbean

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### ABSTRACT

In order to assess the impact of mineral dust on climate and biogeochemistry, it is paramount to identify the sources of dust emission. In this regard, radiogenic isotopes have recently been used successfully for tracing North African dust provenance and its transport across the tropical Atlantic to the Caribbean. Here we present two time series of radiogenic isotopes (Pb, Sr and Nd) in dusts collected at the Cape Verde Islands and Barbados in order to determine the origin of the dust and examine the seasonality of westerly dust outflow from Northern Africa. Aerosol samples were collected daily during two campaigns – February 2012 (winter) and June–July 2013 (summer) – at the Cape Verde Atmospheric Observatory (CVAO) on the island of São Vicente (16.9°N, 24.9°W). A one-year-long time series of aerosols from Barbados (13.16°N, 59.43°W) – a receptor region in the Caribbean – was sampled at a lower, monthly resolution.

Our results resolve a seasonal isotopic signal at Cape Verde shown by daily variations, with a larger radiogenic isotope variability in winter compared to that in summer. This summer signature is also observed over Barbados, indicating similar dust provenance at both locations, despite different sampling years. This constrains the isotope fingerprint of Saharan Air Layer (SAL) dust that is well-mixed during its transport. This result provides unequivocal evidence for a permanent, albeit of variable strength, long-range transport of African dust to the Caribbean and is in full agreement with atmospheric models of North African dust emission and transport across the tropical Atlantic in the SAL.

The seasonal isotopic variability is related to changes in the dust source areas – mainly the Sahara and Sahel regions – that are active all-year-round, albeit with variable contributions in summer versus the winter months. Our results provide little support for much dust contributed from the Bodélé Depression in Chad – the “dustiest” place on Earth – reaching Cape Verde and Barbados during the summer, while contributions during the winter months are likely patchy and minor at most.

Importantly, a short-term isotopic excursion is resolved in the Cape Verde winter record during a dust outbreak on 06–08 February 2012. This features a highly radiogenic Pb and Sr and unradiogenic Nd signature, marking a clear shift in dust provenance relative to that of normal days. As the dust storm waned, continuous gradual changes are observed, reflecting mixing and progressive dilution with dust typical of normal days. These inferences from radiogenic isotope tracers are corroborated by both satellite images (CALIPSO and MODIS) and back-trajectory analyses. The radiogenic isotope fingerprinting of these presently-active North African dust sources, and especially the Saharan Air Layer, will prove invaluable

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in studies of past dust emission from Northern Africa, where imagery and back trajectory analysis are unavailable.

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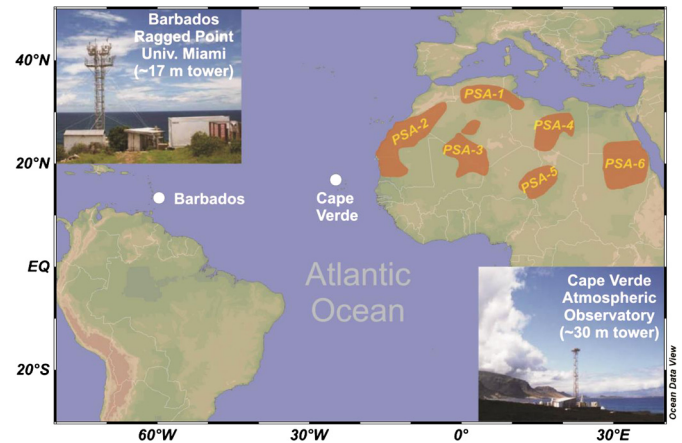
## 1. Introduction

Atmospheric mineral dust has been recognized as an important constituent in the Earth climate system because it impacts atmospheric and oceanic biogeochemical processes (Usher et al., 2003; Shao et al., 2011). The Sahara and Sahel regions in North Africa are of particular importance since they release more than 1000 Tg of mineral dust annually, the largest overall contribution to global dust emissions (Shao et al., 2011). The vast quantities of dust emitted are subsequently transported along three major routes: (1) westward to the tropical Atlantic including over the Gulf of Guinea (Engelstaedter and Washington, 2007; Knippertz and Todd, 2010); (2) northward to Europe (Moreno et al., 2006; Moulin et al., 1998); and (3) eastward to the Middle East via the Mediterranean Sea (Alpert and Ganor, 2001; Prospero et al., 2014). The westward pathway is the dominant outflow (Romero et al., 1999; Schepanski et al., 2009), carrying African dust to the Caribbean Sea and the southern United States during the summer season (Prospero and Lamb, 2003; Prospero and Mayol-Bracero, 2013), then shifting toward northern South America in the winter months (Swap et al., 1992; Koren et al., 2006; Ben-Ami et al., 2010).

African dust export adversely affects the air quality at the receptor regions (Prospero, 1999; Prospero et al., 2014), as well as influencing regional radiative forcing and cloud/cyclone formation over the Atlantic Ocean and the Amazon rainforest (Kaufman et al., 2005; Evan et al., 2006; Martin et al., 2010; Koren et al., 2010). Furthermore, dust acts as a carrier of nutrients, pathogens and disease-spreading spores, which may affect both marine and terrestrial ecosystems upon deposition (Baker et al., 2013; Bristow et al., 2010; Shinn et al., 2000; Swap et al., 1992; Garrison et al., 2003).

Many studies have sought to identify the location of active potential source areas (PSA) of eolian dust emission from within North Africa in order to understand better the dust transport pathways involved (see Engelstaedter et al., 2006). These studies have included satellite observations (Koren et al., 2006; Ridley et al., 2012), modeling approaches (Schepanski et al., 2009; Kim et al., 2014), and field experiments (Haywood et al., 2008; Ansmann et al., 2011). Precise identification of dust source regions and their temporal variations are especially important for validating the parameterization of regional dust emission models. However, remote sensing and modeling approaches do not provide quantitative information on dust concentrations, composition or mineralogy, making it difficult to link an aerosol sample on a filter to a specific dust source.

Attempts to characterize individual sources based upon chemical compositions and clay mineralogy have been summarized by Formenti et al. (2011) and Scheuven et al. (2013). These methods cannot uniquely fingerprint source regions nor identify specific areas of dust emission (see Patey et al., 2015). This is due to the inherent non-uniqueness of mineral dust compositions and clay mineralogy from different source regions. Furthermore, mixing during dust uplift and transport, as well as potential homogenization and grain-size sorting during long-range transport, can “blur” the primary source(s) signature. Such shortcomings can be largely overcome using radiogenic isotope tracers of mineral dust, which have a proven record in numerous provenance studies of both modern and past dust (Abouchami et al., 2013; Grousset and Biscaye, 2005; Kumar et al., 2014; Pourmand et al., 2014; Skonieczny et al., 2011, 2013).



**Fig. 1.** Map showing the location of the sampling site at Sao Vicente Island, Cape Verde, and Barbados in the tropical Atlantic Ocean. The insets show the sampling towers at the sites, on which the samplers were mounted for aerosol collection. Potential source areas (PSA) of dust emissions in North Africa are also shown in the map (adapted from Formenti et al., 2011).

The recent compilation of radiogenic Sr and Nd isotopic datasets of aerosols, sediments and soils from North Africa by Scheuven et al. (2013) provides a reference framework for differentiating source regions of African dust. The Bodélé Depression – a major dust emission source located just north of Lake Chad (e.g. Washington and Todd, 2005; Koren et al., 2006) – has been characterized isotopically (Sr, Nd, and Pb) for the first time by Abouchami et al. (2013) and shown to be very heterogeneous. Subsequent studies combining radiogenic isotopes and air mass back trajectories have confirmed that trans-Atlantic transport of dust takes place from multiple regions in North Africa (Kumar et al., 2014; Pourmand et al., 2014), and has revealed large temporal variability, on weekly to monthly timescales, at the receptor regions (Kumar et al., 2014).

To understand better the seasonality in dust outflow from North Africa and how it relates to dust emitting regions, we present a Sr, Nd and Pb radiogenic isotope time series of African dust collected at the Cape Verde Atmospheric Observatory (CVAO), during a winter (2012) and a summer (2013) season at daily resolution. These data are complemented by a one-year time series of measurements from 2007 at the Barbados receptor region at monthly resolution. Together, these provide an integrated synoptic picture of dust emission and transport across the Atlantic to the Caribbean.

## 2. Materials and methods

### 2.1. Cape Verde sampling

Atmospheric aerosol sampling was undertaken at the Cape Verde Atmospheric Observatory (CVAO), situated on the northeast coast of São Vicente Island (16.9°N, 24.9°W; Fig. 1), about 800 km west of the African coast, off Dakar (Senegal). The site is located on the westward trajectory of the main dust outflow from North Africa to the tropical Atlantic Ocean. More details concerning the sampling site can be found in Fomba et al. (2014).

Bulk aerosols (Total Suspended Particulate, hereafter TSP) and a few PM<sub>10</sub> (particulate matter with aerodynamic diameter <10 μm) samples were collected on top of the 30-m CVAO tower, facing a

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