



# Radiogenic isotopes for deciphering terrigenous input provenance in the western Mediterranean



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## ABSTRACT

Radiogenic isotopic signatures in marine sediments can be used to trace terrigenous source areas and transport mechanisms, which are in turn related to climate variability. To date, most of the published studies using this approach have been focused on eastern Mediterranean sediments. In contrast, we study here the terrigenous input provenance in the westernmost Mediterranean (Alboran Sea basin) by using radiogenic isotope proxies and Nd model ages in a marine record spanning the last 20 ka. Nd, Sr and Pb isotopes, obtained from carbonate-free samples from the <37  $\mu\text{m}$  size fraction, were used to characterize terrigenous variations, including eolian input. Substantial shifts in Pb isotopic signatures throughout the studied time interval reveal a change from North African dominated sources during the glacial period to European dominated sources during the Holocene. Nd and Sr shifts likewise indicate two main short-term changes in sediment provenance, during the last Heinrich event and the early–middle Holocene transition (ca. 8.9 ka cal. BP). Nd model ages over 1.45 Ga also support a contribution of an older component in the terrigenous source, likely Archaean material from the present Senegal region, during both periods. Conversely, terrigenous material mainly shows a dominant provenance from present-day Morocco, Mali, Mauritania, Niger, and Algeria, mixed with material from southern Iberia and southern France. Source variations in the westernmost Mediterranean were mainly driven by fluctuations in wind intensity and fluvial discharges. These fluctuations seem to have been modulated by the African monsoon system further conditioned by the ITCZ migrations and the position of the North Atlantic anticyclone system.

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## 1. Introduction

The composition of terrigenous constituents in sediments of marginal marine basins mainly derives from riverine and eolian inputs, thus reflecting climatic conditions over adjacent continental regions (e.g., Kolla et al., 1979; Jeandel et al., 2007). In the case of the Mediterranean region, its proximity to the Sahara desert makes this area a key location for tracking terrigenous provenance and variations in source areas. In the western Mediterranean, the main sources of terrigenous material are the eolian fraction from arid and semi-arid regions in North Africa, the suspended fluvial particles from the southwestern Europe and North African runoff, and the suspended particulate matter from the Atlantic and Mediterranean waters (Grousset et al., 1988; Bergametti et al., 1989a, 1989b; Lojze-Pilot and Martin, 1996; Molinaroli, 1996; Prospero, 1996; Stumpf et al., 2011). The Western Sahara dry land in North Africa is recognized as a major eolian dust source (Prospero, 1999; Goudie and Middleton, 2001;

Stuut et al., 2009; Prospero and Mayol-Bracero, 2013). Numerical simulations conclude that North Africa is the largest single source of dust on Earth, with up to  $8 \times 10^{14}$  g/a of atmospheric mineral dust, providing 50–70% of total dust emissions (Goudie and Middleton, 2001; Laurent et al., 2008). Extrapolating this figure to the total western Mediterranean basin area (840,000 km<sup>2</sup>) yields an average total atmospheric flux of  $10.9 \pm 0.6 \times 10^{12}$  g/a (Bergametti et al., 1989a, 1989b; Lojze-Pilot et al., 1989). The most important source areas of Saharan dust are located in Western Sahara, Mauritania and Senegal, northern Mali, Atlas Mountains through Morocco, Algeria and Tunisia, central and eastern Libya, western Chad (Bodélé depression), southern Egypt, and northern Sudan (Molinaroli, 1996; Moreno et al., 2006; Laurent et al., 2008; Stuut et al., 2009; Scheuven et al., 2013). Dust source areas over Africa have changed over the past depending on the boundaries of different wind systems, their intensities, and the palaeo-positioning and migrations of the Inter-Tropical Convergence Zone (ITCZ) in response to factors such as orbital-induced summer insolation or changes in cross-equatorial temperature gradients (e.g., Nicholson, 2009; McGee et al., 2014). Furthermore, at the scale of Quaternary climatic oscillations, eolian dust fluxes to the ocean may have been higher during stadial (Greenland Stadial, GS) than interstadial periods (Greenland Interstadial, GI: nomenclature based on INTIMATE group, Lowe et al., 2008), as the

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result of a general southward displacement of the ITCZ during boreal summer (e.g., Reader et al., 1999; Elenga et al., 2000). At present, the relative amount of dust input from the Sahara and Sahelian regions to the western Mediterranean is low and occurs in winter (Bergametti et al., 1989b; Dulac et al., 1992), while in the eastern Mediterranean it is favoured during stronger summer westerlies (Jilbert et al., 2010). The terrigenous particles deflated from the surface depend on several factors including wind speed, atmospheric instability, height of the source area, particle size, particle exposure, soil moisture, vegetative cover, and mineralogical composition (e.g., deMenocal, 1995, 2004; Moreno et al., 2006; Mulitza et al., 2008).

The determination of terrigenous particle provenance, of both eolian or riverine input, in Mediterranean marine sediments has, in general, been based on geochemical and mineralogical data (e.g., Coude-Gaussen et al., 1987; Foucault and Mélières, 2000; Wehausen and Brumsack, 2000; Caquineau et al., 2002; Weldeab et al., 2002a, 2002b, 2003; Díaz-Hernández et al., 2011; Formenti et al., 2011) as well as on remote sensing methods, analysis of surface dust observations, back-trajectory analysis, and the use of mineral tracers (e.g., Goudie and Middleton, 2001; Laurent et al., 2008). Radiogenic isotopes (Sr, Nd, Pb) are likewise powerful tracers for identifying and characterizing source areas of terrigenous material, which may in turn give us additional information about the provenance and transport mechanisms (e.g., Grousset et al., 1988, 1992, 1998; Revel et al., 1996; Tütken et al., 2002; Grousset and Biscaye, 2005; Jullien et al., 2007; Cole et al., 2009; Box et al., 2011; Meyer et al., 2011; Stumpf et al., 2011; Scheuvs et al., 2013).

Previous research in a West–East Mediterranean transect characterized the spatial distribution of detrital flux and terrigenous provenance in Late Pleistocene and Holocene sediments, obtaining as main radiogenic end members Saharan dust and Nile particulate matter (e.g., Krom et al., 1999a, 1999b; Weldeab et al., 2002a, 2002b, 2003; Revel et al., 2010; Box et al., 2011; Blanchet et al., 2013). In the westernmost Mediterranean area, however, less work has focused on the identification of terrigenous provenance and transport patterns. Thus, here we provide a novel and detailed characterization of the terrigenous material and eolian input provenance using radiogenic signatures in marine sediments from a sediment record in the Alboran Sea basin. Previous mineralogical, geochemical and sedimentological analyses from this record have been used to reconstruct the climate variability in terms of atmospheric and oceanic responses as well as to identify eolian input variations since the Last Glacial Maximum (LGM) (Rodrigo-Gámiz et al., 2011, 2014a,b). However, the terrigenous provenance had not been described yet. In this study, we use Sr, Nd, and Pb isotopes and Nd model ages ( $T_{DM}$ ) to constrain for the first time the geographic provenance of terrigenous material and eolian dust, and their transport mechanisms during the last 20 ka.

### 1.1. Radiogenic isotopes as proxies for unravelling chemical weathering or terrigenous source

The variability in radiogenic isotope composition of rocks, which are ultimately the source of the particulate matter suspended and transported by winds or rivers, is essentially the result of chemical fractioning between radioactive parents and radiogenic daughters operated by large-scale geological and ageing processes (Frank, 2002). Consequently, rocks of different ages and provenance in terms of large timescale reservoirs, e.g. mantle vs. crust, have significantly and measurably different isotopic compositions. The isotope composition of the terrigenous material that is the weathering product of the source rocks may coincide or not with that of the source rock, as discussed below.

Nd is a relatively immobile element during weathering and therefore the Nd isotope composition in weathering products is stable during changes from wet to dry periods (e.g., Nesbitt et al., 1980; Dickin, 1997; Braun et al., 1998; Frank, 2002). In contrast, the Sr isotope composition is associated with changes in weathering intensity (Nesbitt et al., 1980; Frank, 2002), and thus radiogenic Sr is

preferentially removed from the source region, leaving a residue with low Sr isotope ratios (Blum and Erel, 1997). During prolonged chemical weathering the radiogenic Sr fraction of a rock is extracted from the source faster than the non-radiogenic Sr fraction, due to the preferential breakdown of Rb-rich phases such as mica and K-feldspar (Nesbitt et al., 1980; Frank, 2002). Therefore the combination of Sr–Nd isotope variations could reflect two different signatures, i.e. changes in the source area, and intense chemical weathering periods involving enhanced rainfall and riverine runoff (Frank, 2002). The extent of chemical weathering is strongly related to prevailing environmental conditions in the source area, such as cover vegetation or aridity. Furthermore, while the Nd isotope signature is unaffected by grain size variations (cf. Goldstein et al., 1984; Grousset et al., 1992), Sr isotopes have shown a relationship with grain size, i.e. Sr isotope ratios increase with decreasing grain size, as a result of the high Rb/Sr in finer sediments (cf. Biscaye and Dasch, 1971; Wehausen and Brumsack, 2000).

Additionally, the Pb isotope composition provides information on source provenances since natural Pb can be transported in the detrital material or in eolian dust for long distances (Grousset et al., 1994; Grousset and Biscaye, 2005; Kylander et al., 2005). However, anthropogenic Pb contamination can overprint the natural Pb isotope signal (Grousset et al., 1994; Kylander et al., 2005). A particular advantage of the Th–U–Pb system is that binary mixtures form straight linear arrays in Pb–Pb isotope space, and deviations from such arrays imply mixtures involving more than two components. Therefore, linear Pb isotope arrays are consistent with binary mixing and imply the existence of multiple and different contributions of Pb sources.

Further information can be obtained from the Nd model ages,  $T_{DM}$ , which provide an isotopic fingerprint of the crustal source, in terms of timing and processes of crust formation (Arndt and Goldstein, 1987). Since the investigated sediments are a mixture of terrigenous material derived from different source areas, i.e. North Africa and South Europe, we expect to obtain “mixed” Nd model ages, which lie between the model ages of the surrounding potential source materials (Fig. 1a).

### 1.2. Present-day climate over the westernmost Mediterranean

Modern climate conditions over the western Mediterranean and northwestern Africa areas are governed by the Azores high-pressure system linked to the North Atlantic climate variability and by African monsoonal dynamics. Summertime climates are usually dry and hot due to the influence of the atmospheric subtropical high-pressure belt (Sumner et al., 2001). During winter the subtropical high shifts to the south, allowing mid-latitude storms to enter the region from the open Atlantic and bringing enhanced amounts of rainfall to the western Mediterranean. This humidity regime in the western Mediterranean is mainly modulated by the North Atlantic Oscillation (NAO) (Hurrell, 1995; Trigo et al., 2002). A high (positive) NAO index causes a more northerly position of the North Atlantic depression, stronger than usual westerlies and warm and wet winters over Northern Europe and dry and cold winters over southern Europe, the Mediterranean and northern Africa (Wanner et al., 2001; Trigo et al., 2002). Conversely, a low (negative) mode of the NAO leads to opposite conditions.

In addition, local climatology of Northern Africa is seasonally modulated by the latitudinal shift of the ITCZ and hence the African monsoon front. The ITCZ directly controls the location of precipitation over North Africa; changes in its position affect how much rainfall occurs and therefore the degree of river runoff. The review by Nicholson (2009) suggests a complex ITCZ pattern, especially for the boreal summer situation. In winter, the equator-ward displacement of the ITCZ (10°N) (Fig. 1a) causes a southward shift of dry subtropical air masses and is associated with the development of strong easterly Saharan Air Layer (SAL) winds, with a northern branch (NSAL). Consequently, a dust plume, usually generated by low-pressure systems, is transported between 15° and 25°N along an E–W axis over the tropical Atlantic Ocean (Holz, 2004) and over the Mediterranean (Moulin et al., 1997).

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