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Lead isotopic compositions in the EPICA Dome C ice core and Southern Hemisphere Potential Source Areas☆

P. Vallelonga [a,b,*], P. Gabrielli [c], E. Balliana [d,e], A. Wegner [f], B. Delmonte [g], C. Turetta [a], G. Burton [b], F. Vanhaecke [d], K.J.R. Rosman [b,1], S. Hong [h], C.F. Boutron [i,j], P. Cescon [a,e], C. Barbante [a,e]

[a] Institute for the Dynamics of Environmental Processes – CNR, Calle Larga Santa Marta 2137, I-30123 Venice, Italy
[b] Department of Imaging and Applied Physics, Curtin University of Technology, GPO Box U1987, Perth, WA 6845, Australia
[c] School of Earth Sciences and Byrd Polar Research Center, The Ohio State University, 108 Scott Hall, 1090 Carmack Road, Columbus, OH 43210-1002, USA
[d] Department of Analytical Chemistry, Ghent University, Krijgslaan 281-S12, B-9000 Ghent, Belgium
[e] Department of Environmental Sciences, University of Venice Ca' Foscari, Calle Larga Santa Marta 2137, I-30123 Venice, Italy
[f] Alfred-Wegener-Institut, Am Alten Hafen 26, D-27568 Bremerhaven, Germany
[g] Dipartimento di Scienze dell'Ambiente e del Territorio, Universitá degli Studi di Milano-Bicocca, Piazza della Scienza 1, 20126, Milan, Italy
[h] Korea Polar Research Institute (KOPRI), 806 Get-Pearl Tower, Songdo Technopark, 7-50 Songdo-dong, Yunsu-ku, Incheon, 406-840, Korea
[i] Laboratoire de Glaciologie et Géophysique de l'Environnement du C.N.R.S., 54 rue Molière – Domaine Universitaire – BP 96 – 38402 St Martin d'Hères Cedex, France
[j] Université Joseph Fourier de Grenoble (Institut Universitaire de France), B.P. 68, 38041 Grenoble, France

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ABSTRACT

A record of Pb isotopic compositions and Pb and Ba concentrations are presented for the EPICA Dome C ice core covering the past 220 ky, indicating the characteristics of dust and volcanic Pb deposition in central East Antarctica. Lead isotopic compositions are also reported in a suite of soil and loess samples from the Southern Hemisphere (Australia, Southern Africa, Southern South America, New Zealand, Antarctica) in order to evaluate the provenance of dust present in Antarctic ice. Lead isotopic compositions in Dome C ice support the contention that Southern South America was an important source of dust in Antarctica during the last two glacial maxima, and furthermore suggest occasional dust contributions from local Antarctic sources. The isotopic signature of Pb in Antarctic ice is altered by the presence of volcanic Pb, inhibiting the evaluation of glacial–interglacial changes in dust sources and the evaluation of Australia as a source of dust to Antarctica. Consequently, an accurate evaluation of the predominant source(s) of Antarctic dust can only be obtained from glacial maxima, when dust-Pb concentrations were greatest. These data confirm that volcanic Pb is present throughout Antarctica and is emitted in a physical phase that is free from Ba, while dust Pb is transported within a matrix containing Ba and other crustal elements.

1. Introduction

Dust is an active component of the Earth's climate system, both contributing to and responding to changes in radiative forcing and ocean biogeochemistry (Mahowald et al., 2005). The polar ice sheets are an ideal archive for monitoring variations in dust fluxes and other climate parameters, and have been crucial to understanding the role of dust in climate variability (Fischer et al., 2007). It has recently been suggested that variations in Antarctic dust fluxes are primarily driven by strengthened Aeolian deflation in source regions and weakening of the hydrological cycle rather than variations in atmospheric transport parameters or moderations in meridional transport (Lambert et al., 2008).

☆ This paper is dedicated to the memory of our friend and colleague Professor Kevin J.R. Rosman who, among other achievements, pioneered the accurate determination of lead isotopic compositions in polar snow and ice.

* Corresponding author. Tel.: +39 041 2348504; fax: +39 041 2348628.
E-mail addresses: vallelonga@unive.it (P. Vallelonga), gabrielli.1@osu.edu (P. Gabrielli), eleonora.balliana@ugent.be (E. Balliana), anna.wegner@awi.de (A. Wegner), barbara.delmonte@unimib.it (B. Delmonte), clara.turetta@idpa.cnr.it (C. Turetta), g.burton@curtin.edu.au (G. Burton), frank.vanhaecke@ugent.be (F. Vanhaecke), smhong@kopri.re.kr (S. Hong), boutron@lgge.obs.ujf-grenoble.fr (C.F. Boutron), cescon@unive.it (P. Cescon), barbante@unive.it (C. Barbante).

[1] Deceased.

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Grousset and co-workers have demonstrated the utility and selectivity of Sr and Nd isotopic systems for investigation of dust provenance in ice cores (Grousset and Biscaye, 2005). Initial studies required kg-size samples, limiting analytical resolution and sample availability to a few glacial samples, but ongoing development of the analytical technique has seen improvements in sample resolution and recently low-concentration interglacial samples have been analysed (Delmonte et al., 2007).

In parallel, the isotopic signature of dust from Potential Source Areas (PSAs) has been documented. The recent work of Gaiero (2007) reported Sr and Nd isotopic composition for PSA in Southern South America (SSA) indicating the importance of considering high-latitude areas such as Patagonia as well as low-latitude sources located at high altitude such as the Puna-Altiplano plateau in the Andean Cordillera. Revel-Rolland et al. (2006) reported an extensive data set of Sr and Nd isotopic signatures in East Australian PSA samples, suggesting the presence of a non-trivial Australian dust contribution to Antarctica during interglacial climate phases. They suggested a 10–20% Australian dust contribution during glacial phases and up to 50% during interglacial phases. The changing proportions of dust sources was attributed to changes in primary production of dust in SSA during glacial climate phases resulting from glacial and periglacial processes related to the growth and recession cycles of the Patagonian ice cap.

The use of Pb isotopes for dust provenance has been limited on account of the extreme contamination controls required to accurately determine Pb isotopic ratios in pg-level quantities, as well as a dearth of Pb isotope data for PSAs. Initial studies of Pb isotopes in Antarctic ice (Rosman et al., 1994) and seawater (Flegal et al., 1993) reported the extent and origins of industrial pollution in the Antarctic environment. Vallelonga et al. (2002a,b) demonstrated that industrial Pb emissions were efficiently transported to Antarctica from Australia from 1880 AD on, with the influence of leaded gasoline emissions from South America evident since the 1960s. This work demonstrated the efficacy of fine particle transport mechanisms from Australia to coastal Antarctica, and established the extremely low concentrations of Pb present in Antarctic ice during the Holocene of $\sim$0.3 pg/g (0.3×10^{-12} g/g).

Determination of the provenance of Antarctic dust using Pb isotopes has been hampered by the presence of volcanic Pb in Antarctica, which has a different isotopic composition to that of dust Pb. Matsumoto and Hinkley (2001) demonstrated the overwhelming presence of highly radiogenic volcanic Pb in the Taylor Dome ice core, located west of the Transantarctic mountains, with volcanic sources accounting for $\sim$60% of total Pb in Taylor Dome ice during the glacial phases and $\sim$95% during the Holocene. Determinations of volcanic Pb inputs are usually based on Pb/Ba or Pb/Al ratios measured in ice samples, with Ba or Al used as a continental dust proxy, and the dust-Pb contribution calculated using an estimate of the ratios of the elements in the upper continental crust (e.g. Wedepohl, 1995). Data from other locations in Antarctica indicate comparable or lower volcanic Pb contributions during the Holocene: 80–90% volcanic Pb contributions to Law Dome (Vallelonga et al., 2002a,b) and Victoria Land (Van de Velde et al., 2005), 30–80% volcanic Pb contributions to Coats Land (Planchon et al., 2003), $\sim$20% volcanic Pb contributions to Vostok (Hong et al., 2003) and $\sim$15% volcanic Pb contributions to Dome C (Vallelonga et al., 2005). Hinkley (2007) demonstrated that these volcanic Pb contributions converge on a radiogenic Pb isotopic composition of $^{206}Pb/^{207}Pb \sim 1.26$, which corresponds to the Pb isotopic signatures of ocean island volcanoes as well as Antarctic volcanic provinces, including Mount Erebus.

The availability of data for PSAs has been another limitation to dust provenance studies using Pb isotopes. Lead isotopic compositions have been reported for South Atlantic and South Pacific Ocean pelagic sediments (Chow and Patterson, 1962) but no land-based loess or soil data were available before this study. Geochronological studies of volcanic provinces of Antarctica [Erebus: Sun and Hanson (1975), Sims et al. (2008); Marie Byrd Land: Hart et al. (1997); Antarctic Peninsula: Hole et al. (1993)] and the South Sandwich Islands (Barreiro, 1983) ensure that the volcanic Pb component is already well defined. These studies support the radiogenic signature of Antarctic volcanic emissions indicated by Vallelonga et al. (2002a,b) and Hinkley (2007). Rosman et al. (1998) also identified rocks from a Th-rich province in Enderby Land with matching Pb isotopic compositions in Law Dome ice.

Recent analysis of Sr and Nd isotopic compositions in targeted locations of Southern South America and Australia have greatly stimulated the interpretation of Antarctic ice core data. This work seeks to provide a similar stimulus for the analysis of Antarctic Pb isotope compositions, by presenting Pb isotopic compositions in a set of targeted Australian soil and loess samples in arid locations of identified dust production and prolonged aridity during the Quaternary. Furthermore, Pb isotopes are reported for selected samples previously analysed for Sr and Nd by Delmonte et al. (2004) for comparison with existing proxies of PSAs, such as pelagic sediments and volcanic lavas. New data from the EPICA Dome C ice core are also presented, to improve the statistical reliability of dust provenance evaluations. This work contributes to evaluations of the provenance of Antarctic aerosol, distinguishing between dust and volcanic contributions, and provides an important data set for interpretation of Pb isotopes in Antarctic ice and snow in future studies.

2. Regional setting

2.1. Ice core samples

All ice core samples presented here are from the EPICA Dome C (hereafter EDC) core drilled between 1996 and 2005. The location and site characteristics of EDC have already been extensively described (EPICA community members, 2004). Sample dating was assigned using the EDC3 chronology of Parrenin et al. (2007) with paleotemperature information provided by the high-resolution record of deuterium/hydrogen ratios (represented here as δD) reported by Jouzel et al. (2007). A preliminary data set of 30 EDC samples (Vallelonga et al., 2005) has here been extended to 74 samples covering the period 2–217 kyr BP (86.6–2193.4 m depth). Some of the EDC samples previously reported have also been remeasured to improve the precision of the Pb isotopic compositions (sample depths: 229.1 m, 432.6 m, 489.0 m, 598.1 m, 709.0 m, 1093.1 m, 1643.1 m, 2094.4 m).

2.2. Australian PSA samples

PSA samples from Australia were collected in July 2007 from various loess and soil deposits for which chronologies and geomorphological descriptions have already been reported. The sites sampled include Lake Mungo dunefield (Kershaw and Nanson, 1993), Brachina Gorge (Glasby et al., 2007), Lake Eyre (Magee et al., 2004), Strzelecki desert (Fitzsimmons et al., 2007) and Innamincka/Tilcha Waterhole (Nanson et al., 2008). These sites represent locations of present dust production or loess deposits, which can be considered isotopically representative of dust produced in previous glacial periods.

2.3. Other PSA samples

Lead isotopic compositions were determined in a selection of soil and loess samples previously reported by Delmonte et al. (2004). These samples derive from locations within SSA (in particular from