



# The distribution of lead concentrations and isotope compositions in the eastern Tropical Atlantic Ocean

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## Abstract

Anthropogenic emissions have dominated marine Pb sources during the past century. Here we present Pb concentrations and isotope compositions for ocean depth profiles collected in the eastern Tropical Atlantic Ocean (GEOTRACES section GA06), to trace the transfer of anthropogenic Pb into the ocean interior. Variations in Pb concentration and isotope composition were associated with changes in hydrography. Water masses ventilated in the southern hemisphere generally featured lower  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{208}\text{Pb}/^{207}\text{Pb}$  ratios than those ventilated in the northern hemisphere, in accordance with Pb isotope data of historic anthropogenic Pb emissions. The distributions of Pb concentrations and isotope compositions in northern sourced waters were consistent with differences in their ventilation timescales. For example, a Pb concentration maximum at intermediate depth (600–900 m,  $35 \text{ pmol kg}^{-1}$ ) in waters sourced from the Irminger/Labrador Seas, is associated with Pb isotope compositions ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.1818\text{--}1.1824$ ,  $^{208}\text{Pb}/^{207}\text{Pb} = 2.4472\text{--}2.4483$ ) indicative of northern hemispheric emissions during the 1950s and 1960s close to peak leaded petrol usage, and a transit time of  $\sim 50\text{--}60$  years. In contrast, North Atlantic Deep Water (2000–4000 m water depth) featured lower Pb concentrations and isotope compositions ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.1762\text{--}1.184$ ,  $^{208}\text{Pb}/^{207}\text{Pb} = 2.4482\text{--}2.4545$ ) indicative of northern hemispheric emissions during the 1910s and 1930s and a transit time of  $\sim 80\text{--}100$  years. This supports the notion that transient anthropogenic Pb inputs are predominantly transferred into the ocean interior by water mass transport. However, the interpretation of Pb concentration and isotope composition distributions in terms of ventilation timescales and pathways is complicated by (1) the chemical reactivity of Pb in the ocean, and (2) mixing of waters ventilated during different time periods. The complex effects of water mass mixing on Pb distributions is particularly apparent in seawater in the Tropical Atlantic Ocean which is ventilated from the southern hemisphere. In particular, South Atlantic Central Water and Antarctic Intermediate Water were dominated by anthropogenic Pb emitted during the last 50–100 years, despite estimates of much older average ventilation ages in this region.

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## 1. INTRODUCTION

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During the past century, emissions from anthropogenic activities completely overwhelmed oceanic Pb inventories (Boyle et al., 2014). Of particular importance was the use

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of leaded petrol along with high temperature industrial processes such as metal smelting, coal combustion and waste incineration (Nriagu and Pacyna, 1988; Pacyna and Pacyna, 2001). Radiogenic Pb isotope compositions can provide insight into different sources of Pb to environmental systems. Notably, the isotope compositions of anthropogenic Pb emissions exhibit regional variations, reflecting the distinct Pb ores used to supply industrial and automotive activities in different economic areas (e.g. Bollhöfer and Rosman, 2001). This has resulted in spatial variability in the Pb isotope composition of ocean surface waters, which can be used to assess the relative contribution of anthropogenic and natural Pb inputs from different regional sources (Véron et al., 1994, 1998; Hamelin et al., 1997; Weiss et al., 2003; Noble et al., 2015; Paul et al., 2015a; Bridgestock et al., 2016).

Both the magnitude and isotope composition of anthropogenic Pb fluxes to the oceans have changed over time in response to known changes in anthropogenic activities (Boyle et al., 1986; Shen and Boyle, 1987; Véron et al., 1993; Wu and Boyle, 1997; Desenfant et al., 2006; Kelly et al., 2009). For example, since the 1920s Pb concentrations in North Atlantic surface waters have closely tracked the utilization of leaded petrol in the surrounding regions with a peak in the 1970s (Kelly et al., 2009). Accompanying temporal changes in the Pb isotope composition of Atlantic surface waters have been attributed to changes in the relative magnitude of Pb emissions from different areas (i.e. the US and Europe) as well as changes in the market share of particular Pb ores used within regions (Wu and Boyle, 1997; Véron et al., 1998; Hurst, 2002; Kelly et al., 2009).

These transient Pb inputs were subsequently transferred into the ocean interior predominantly by ocean circulation, as highlighted by time series measurements of Pb concentrations and isotope compositions throughout the water column at the Bermuda Time Series station conducted since 1979 (Chow and Patterson, 1966; Schaule and Patterson, 1983; Boyle et al., 1986; Shen and Boyle, 1988; Sherrell and Boyle, 1992; Véron et al., 1993, 1998; Lee et al., 2011; Boyle et al., 2012; Noble et al., 2015). The distribution of Pb concentrations and isotope compositions in the ocean interior can therefore provide insights into ventilation pathways and timescales, complementary to other anthropogenic transient tracers, such as CFCs, SF<sub>6</sub> and <sup>3</sup>H (e.g. Doney et al., 1997; Fine, 2011). However, Pb concentrations are not conserved during transport by ocean circulation due to removal by particle scavenging (Boyle et al., 1986; Shen and Boyle, 1988). Moreover, equilibrium exchange between adsorbed particulate and dissolved Pb may modify the isotope composition of seawater (Wu et al., 2010; Chen et al., 2016; Zurbrick et al., 2017). Finally, the mixing of waters ventilated at different times, featuring different Pb concentrations and isotope compositions, will complicate interpretations of ventilations pathways and timescales in a similar manner to the ‘non-linear’ mixing effects imposed on other transient tracers (e.g. Doney et al., 1997; Lee et al., 2017).

Unraveling the roles of transient Pb inputs, advection, mixing and particulate transport processes in setting the observed distributions of Pb in the ocean presents signifi-

cant challenges. Efforts to improve the understanding of the processes controlling the cycling of Pb in the ocean are currently hindered by the sparse coverage of Pb concentration and isotope composition data, both spatially and temporally. In addition, the full potential of isotope variations to trace different sources of Pb to the ocean has yet to be realized. Studies tracing environmental Pb sources have typically focused on the relative abundances of <sup>206</sup>Pb, <sup>207</sup>Pb and <sup>208</sup>Pb, whilst neglecting the use of <sup>204</sup>Pb (e.g. Weiss et al., 2003). This is due to analytical difficulties in the precise determination of Pb isotope ratios featuring <sup>204</sup>Pb, which has a low relative abundance of about 1% (Komárek et al., 2008). Hence, source apportionment is typically conducted using only two of three potential independently varying isotope ratios. Recent methodological developments now facilitate the precise measurement of Pb isotope ratios featuring <sup>204</sup>Pb in seawater, and thus the ability to fully utilize the potential of Pb isotope variations for source assessment (Paul et al., 2015b).

Here we present Pb concentrations and isotope compositions for seawater depth profiles and Pb concentrations of suspended particulate material collected in the eastern Tropical Atlantic Ocean (Fig. 1). Notably, the Pb isotope data include high precision results for ratios featuring <sup>204</sup>Pb. These results are used to assess the controls on the distribution of Pb across a hydro-dynamically complex region of the ocean, transecting water masses ventilated in the northern and southern hemispheres and significant gradients in surface Pb inputs from natural and anthropogenic sources (Bridgestock et al., 2016).

## 2. SAMPLES AND ANALYTICAL TECHNIQUES

Samples were collected during the GEOTRACES GA06 section cruise on the RRS Discovery (D361; 7th February –

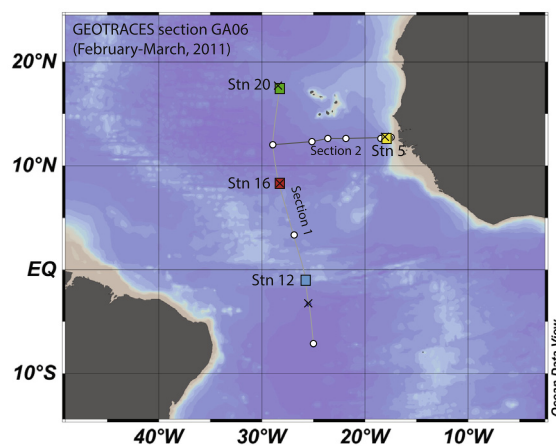


Fig. 1. Locations of samples collected during the GEOTRACES GA06 cruise in 2011. Colored squares show the location of depth profiles collected for Pb concentration and isotope composition analyses, crosses the locations of surface water samples collected close to each depth profile (Bridgestock et al., 2016), and open circles the location of depth profiles of particulate samples. Figure produced using Ocean Data View (Schlitzer, 2015). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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