

Intercomparison of dissolved iron isotope profiles from reoccupation of three GEOTRACES stations in the Atlantic Ocean



T.M. Conway^{a,b,*}, S.G. John^{b,1}, F. Lacan^c

^a Department of Earth Sciences, ETH Zürich, Zürich, Switzerland

^b Department of Earth and Ocean Sciences, University of South Carolina, Columbia, SC, United States

^c LEGOS, Université de Toulouse, CNES, CNRS, IRD, UPS, Toulouse, France

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ABSTRACT

Intercomparison of trace metal data is a key aspect of the International GEOTRACES program, allowing data from multiple laboratories and countries to be combined to produce high-resolution datasets for the oceans. The use of crossover stations by the GEOTRACES program provides the opportunity both for comparison of analytical techniques and assessment of temporal variability in the cycling of trace metals such as iron (Fe). Here, we present the first comparison of dissolved Fe stable isotope ratio ($\delta^{56}\text{Fe}$; relative to IRMM-014) profiles in the oceans, from reoccupations of three locations in the Atlantic Ocean; (1) the Bermuda Atlantic Time Series station (31.75°N 64.17°W) during the US GEOTRACES IC1 cruise (June 2008) and the US GEOTRACES GA03 cruise (Nov. 2011); (2) the Tenatso Time Series station near Cape Verde (17.4°N 24.5°W) during the U.S GEOTRACES GA03 cruises (2010; 2011), and (3) a station in the Cape Basin close to South West Africa (31.1–31.4°S 36.5°W) during the French GEOTRACES GIPY4 Bonus Good Hope Cruise (Feb. 2008) and the UK GEOTRACES GA10 D357 cruise (Oct. 2010). These datasets provided us with the opportunity not only to compare sampling and analysis techniques by two different laboratories (USC and LEGOS), but also the temporal variability of $\delta^{56}\text{Fe}$ at these locations on a 1–3 year timescale. We found that a good agreement between data and profiles generated by different laboratories does allow assessment of temporal variation of $\delta^{56}\text{Fe}$ in the water column, as well as spatial variability and synthesis of datasets from different regions of the ocean. In fact, comparison of $\delta^{56}\text{Fe}$ at the three locations in this study demonstrates a remarkable consistency between the shapes of ocean $\delta^{56}\text{Fe}$ profiles measured 1–3 years apart, pointing to the overall stability of Fe cycling at all three locations on these timescales, despite the expected dynamic nature of the Fe cycle. This consistency is highlighted by strong agreement in $\delta^{56}\text{Fe}$ throughout the whole water-column at Bermuda, and in waters deeper than 500 m in the Cape Basin, which suggests that different water masses may carry distinct $\delta^{56}\text{Fe}$ signatures. In contrast to these stable $\delta^{56}\text{Fe}$, we did observe apparent temporal variability in $\delta^{56}\text{Fe}$ between cruises at other locations and in surface waters, both throughout the water column at Cape Verde, and in Agulhas-leakage influenced surface waters in the Cape Basin. Such temporal variability may thus provide information about changes in internal Fe cycling or external Fe sources on these timescales. Overall, this study highlights the usefulness of repeat $\delta^{56}\text{Fe}$ measurements to provide information on the variability of Fe cycling throughout the oceans.

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1. Introduction

Seawater dissolved iron (Fe) is an important limiting nutrient for phytoplankton over much of the surface oceans (Moore et al., 2001). Understanding the global distribution and biogeochemical cycling of Fe is a central aim of the international GEOTRACES program which seeks to measure global distributions of a range of trace elements and

their isotopes. Fe concentrations are a GEOTRACES key parameter, meaning their analysis is required on all GEOTRACES cruises (Anderson and Henderson, 2005; Henderson et al., 2007). Fe stable isotopes ($\delta^{56}\text{Fe}$) have also been measured on several GEOTRACES cruises, although they are not a key parameter. The resulting ocean sections of dissolved Fe concentration from the Atlantic, Indian and Pacific Oceans, and similar sections in progress, are beginning to provide new insights into the marine sources, sinks and cycling of Fe at both the regional and global scale throughout the oceans (e.g. Conway and John, 2014a; Klunder et al., 2012; Nishioka et al., 2013; Resing et al., 2015; Rijkenberg et al., 2014; Saito et al., 2013).

GEOTRACES cruises all follow internationally agreed methods for clean sampling and handling (The GEOTRACES Cookbook; Cutter et al.,

* Corresponding author at: Department of Earth Sciences, ETH Zürich, Zürich, Switzerland.

E-mail address: conway.tm@gmail.com (T.M. Conway).

¹ Department of Earth Sciences, University of Southern California, Los Angeles, USA (present address).

2010, 2014), meaning that datasets can be collated to form regional and global pictures of the distribution of trace metals and their isotopes in the ocean. Recently, such data has been combined to form the GEOTRACES Intermediate Data Product and the GEOTRACES eAtlas (Mawji et al., 2015; Schlitzer, 2015). To ensure compatibility of datasets from different groups and nations, given the range of collection and analytical techniques deployed on different cruises, it is essential to carry out intercomparison of data collected from the same location and/or in the same samples by multiple groups. Accordingly, intercomparison is an important aspect of the GEOTRACES program, with the recommended reporting of measurements of SAFe reference standards for dissolved trace metal concentrations, the use of cross-over stations for comparison between different GEOTRACES cruises (see Fig. 1), and a number of studies comparing sampling systems (Cutter, 2013). Intercomparison of trace metal concentrations and stable isotope ratios was a goal of two US GEOTRACES cruises in the Atlantic (IC1, June 2008) and North Pacific (IC2, May 2009) Oceans (Boyle et al., 2012; Cutter and Bruland, 2012; Cutter, 2013; and others), and recent work has compared the clean sampling system of the Dutch and US GEOTRACES programs as well as different analytical techniques (Middag et al., 2015).

The GEOTRACES program and similar cruises have also facilitated the application of dissolved Fe isotope ratios ($\delta^{56}\text{Fe}$) as a useful parameter for understanding the marine Fe cycle. Although a challenging measurement, due to the low concentration of Fe in seawater and the difficulties of analysis, advancement in chemical techniques and high-resolution mass spectrometry has allowed a number of groups to develop methods to measure seawater-dissolved $\delta^{56}\text{Fe}$ over the full range of observed oceanic dissolved Fe concentrations (~ 0.02 to >2 nmol kg^{-1}). These methods have utilized either NTA or Nobias PA-1 chelating resins (Boyle et al., 2012; Conway et al., 2013a; John and Adkins, 2010; Lacan et al., 2010, 2008; Rouxel and Auro, 2010), extraction with dithiocarbamate (Ellwood et al., 2014) or co-precipitation with magnesium (de

Jong et al., 2007), followed by analysis with multicollector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS). Such methods have to date led to the publication of dissolved $\delta^{56}\text{Fe}$ depth profiles from a number of ocean basins (e.g. Chever et al., 2015; John and Adkins, 2012; John et al., 2012; Labatut et al., 2014; Lacan et al., 2008; Radic et al., 2011; Staubwasser et al., 2013), and a high resolution ocean section of dissolved $\delta^{56}\text{Fe}$ across the North Atlantic (Conway and John, 2014a). Dissolved $\delta^{56}\text{Fe}$ measurements were also part of the focus of a previous intercomparison effort utilizing seawater collected from the Bermuda Atlantic Time Series onboard the US GEOTRACES 1C1 cruise in 2008, where four different laboratories demonstrated strong agreement on both surface and deep water samples at concentrations of 0.4 and 0.8 nmol kg^{-1} (Boyle et al., 2012; see Fig. 2c–d).

Different marine sources of Fe have different $\delta^{56}\text{Fe}$ signatures, and these isotope signatures have been used to trace Fe as it mixes through the oceanic water column, including non-reductive sediment dissolution (Labatut et al., 2014; Radic et al., 2011), reductive sediment dissolution (Chever et al., 2015; John et al., 2012), and Fe from multiple sources (Conway and John, 2014a). As such, studies of seawater-dissolved $\delta^{56}\text{Fe}$ show promise for understanding the cycling and differing sources of Fe to the ocean; however, application of this tracer is still limited to a small number of laboratories. Thus, spatial coverage of $\delta^{56}\text{Fe}$ measurements throughout the oceans remains sparse. Additionally, although changing conditions over time are important to our understanding of changes in Fe sources and cycling, logistics and cost have meant that repeat-sampling to assess temporal variability has rarely been carried out. However, reoccupation of crossover stations as part of the GEOTRACES program provides us with the opportunity to begin to address this lack of knowledge of variability, as well as to compare sampling and analysis by different groups.

Here we compare measured Fe and $\delta^{56}\text{Fe}$ profiles from three locations in the Atlantic Ocean (Fig. 1): 1) reoccupation of a GEOTRACES

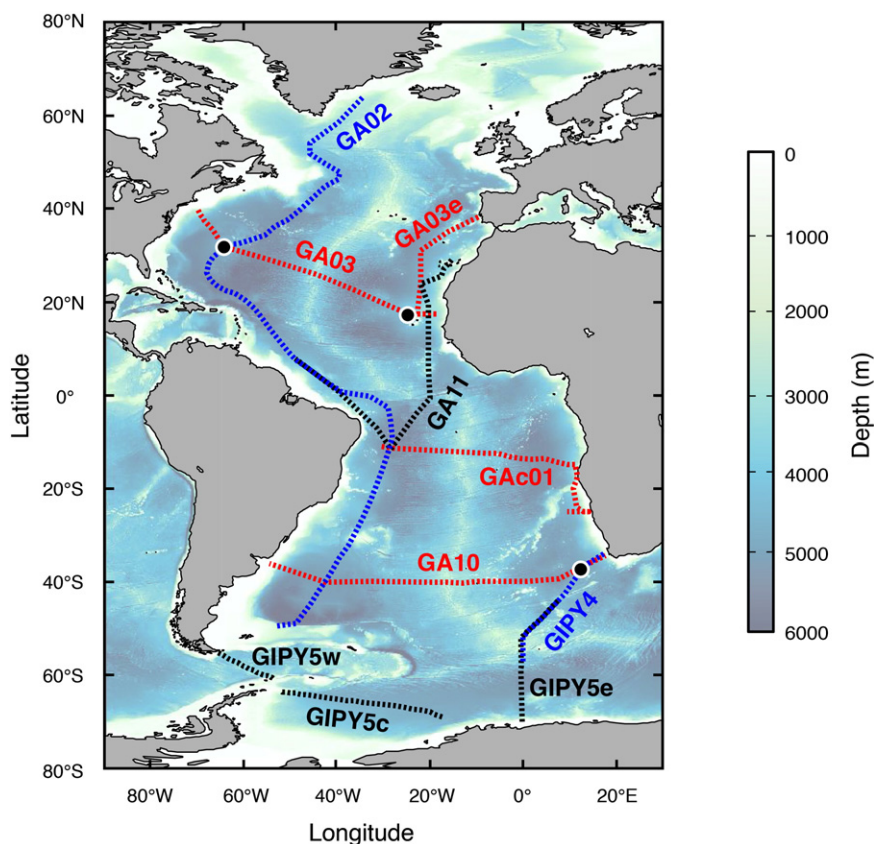


Fig. 1. Atlantic Ocean showing GEOTRACES crossover stations and locations described in this study. Cruises that included in the GEOTRACES Intermediate data product 2014 are shown for reference, based on eGEOTRACES graphics (Mawji et al., 2015; Schlitzer, 2015). The three locations sampled in this study are shown as black dots.

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