PERSPECTIVE

GEOTRACES: IRONING OUT THE DETAILS OF THE OCEANIC IRON SOURCES?

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ABSTRACT. Dissolved iron (dFe) is an essential micronutrient for phytoplankton, with vanishingly low oceanic dissolved concentrations (pico- to nanomoles per kg) known to limit growth—and thus influence primary productivity and carbon cycling—over much of the surface ocean. However, because of the considerable challenges associated with contamination-free sample collection and accurate analysis of such low dFe concentrations, the first reliable dFe measurements came only in the 1980s. Further, by 2003, despite several decades of research, there were only ~25 full-depth oceanic dFe profiles worldwide, with dust considered to be the main oceanic dFe source. Since 2008, facilitated by the extensive field campaign and rigorous intercalibration of the international GEOTRACES program, there has been an "explosion" in the availability of oceanic dFe data, with hundreds of profiles now available. Concurrently, there has been a paradigm shift to a view of the marine Fe cycle where multiple sources contribute, and some forms of dFe can be transported great distances through the intermediate and deep ocean. Here, we showcase the GEOTRACES dFe datasets across the different ocean basins, synthesize our current multi-source view of the oceanic Fe cycle, spotlight sediments as an important dFe source, and look to future directions for constraining oceanic dFe boundary exchange.

MEASURING Fe IN THE OCEAN: A HISTORICAL PERSPECTIVE

Although our understanding of many chemical tracers in the ocean advanced significantly with the GEOSECS program of the 1970s, there remained significant challenges to contamination-free sample collection, filtration, and analysis of sub-nanomolar (10^{-9} mol kg^{-1}) concentrations of dissolved iron in seawater (dFe; here operationally defined as what will pass through a 0.2 or 0.4 micron pore size filter) so that the first accurate oceanic dFe water column profiles date only to the early 1980s (Landing and Bruland, 1981; Gordon et al., 1982). These studies were followed by shipboard incubations showing that sub-nanomolar dFe concentrations limit primary productivity over HNLC (high-nutrient, lower-than-expected chlorophyll) regions of the surface ocean, where major nutrients are underutilized by phytoplankton (Martin and Fitzwater, 1988; Martin et al., 1990a, 1990b). Such findings led to the "Iron Hypothesis" that postulated Fe's role in influencing global climate change and a large body of research, including surface ocean Fe fertilization experiments (Martin, 1990; De Baar et al., 2005; Boyd et al., 2007).

By 1997, a synthesis of available oceanic dFe measurements established a paradigm which asserted that the main source of Fe to the ocean was dissolution of Fe-bearing aeolian dust, and that deep-water dFe concentrations (away from proximal sources) were buffered around 0.6 nmol L^{-1} by complexation with organic ligands (Johnson et al., 1997; Tagliabue et al., 2017). Fe profiles were characterized as "nutrient" type, with depleted surface concentrations due to biological uptake of Fe and subsequent release from particles at depth, or as "hybrid," which also accounted for the dust source and the competing effects of particle scavenging and organic complexation (Bruland and Lohan, 2003; Boyd and Ellwood, 2010; Conway and Middag, in press). In this view, surface dFe concentrations would be elevated only in upwelled deep waters or in surface waters that had received high atmospheric dust fluxes or were very close to other terrestrial Fe sources. The effects of dust addition were later elegantly demonstrated by observations of a dramatic shift in dFe concentrations from 0.1 to 2 nmol kg^{-1} between winter and summer in the subtropical North Atlantic, a region that receives large Saharan dust fluxes only in summer (Sedwick et al., 2005). Fe released from hydrothermal "black smoker" vents was typically thought to be lost to sediments close to the vent sources via precipitation of sulfide and oxide minerals (German et al., 1991).

However, by the birth of the international GEOTRACES endeavor (during discussions in 2003–2004; Jeandel, 2024, in this issue), and despite demonstrable high-quality dFe measurements from multiple international groups from different oceanic regions (Johnson et al., 2007), dFe data were geographically limited to just ~25 depth profiles deeper than 2,000 m (GEOTRACES Planning Group, 2006; Anderson et al., 2014). This paucity of dFe data stymied a complete understanding of much of the marine Fe cycle—especially understanding of the roles of deep Fe sources such as hydrothermal vents and marine sediments in influencing dFe distributions—and it provided impetus for the establishment of the international GEOTRACES program that named dFe as a "key parameter" to be measured on all GEOTRACES cruises. GEOTRACES aimed to "determine global ocean distributions of selected trace elements and isotopes (TEIs)—including their concentrations, chemical speciations, and physical forms—and to evaluate the sources, sinks, and internal cycling of these species to characterize more completely the physical, chemical and biological processes regulating their distributions" (GEOTRACES Planning Group, 2006). As is described later in this article, the knowledge gained from GEOTRACES dFe distributions has led to a shift away from the dust-focused paradigm of the 1990s to a new paradigm wherein multiple boundary sources influence open ocean dFe distributions, and dust inputs are largely restricted to dusty regions of the globe (Tagliabue et al., 2014, 2017).

Our intention here is not to review the entire field of marine Fe research, nor to explore global biogeochemical Fe models, Fe speciation and complexation, the role of internal cycling processes, or marine particles, because this has been aptly done by others (e.g., Boyd and Ellwood, 2010; Tagliabue et al., 2017), or is well covered by other articles in this special issue (Anderson, 2024, in this issue; Twining, 2024; Tagliabue and Weber, 2024; Whitby et al., 2024). Instead, we focus on showcasing the breadth of the GEOTRACES dFe datasets that are publicly available in the latest GEOTRACES data product (IDP2021v2; GEOTRACES Intermediate Data Product Group, 2023) and discuss how our view of the importance and pervasive nature of (non-dust) boundary sources of Fe to the ocean has changed dramatically with the availability of new, high-resolution, geographically distributed dFe and other oceanic TEI datasets since the birth of GEOTRACES.

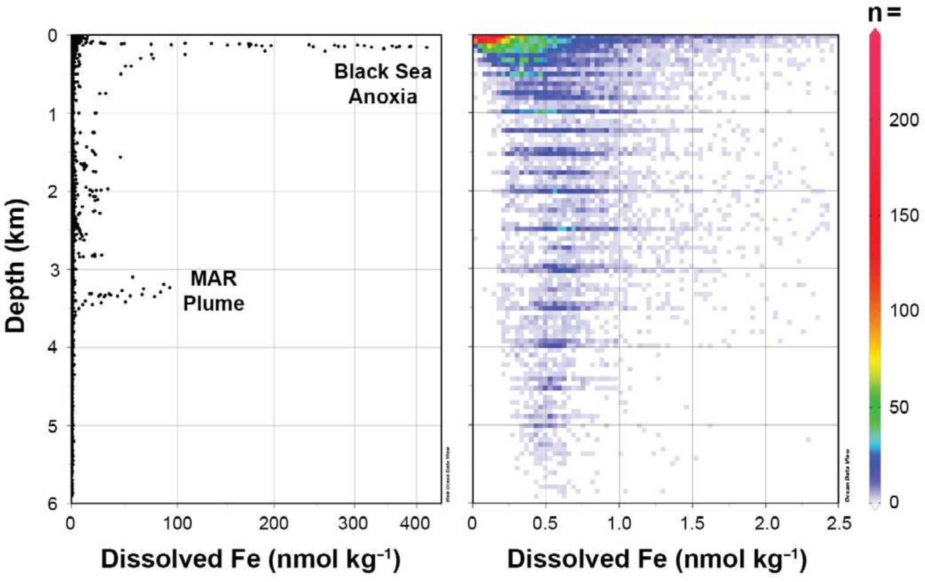

FIGURE 1. Distribution of all dissolved Fe concentration data included in the GEOTRACES Data Product 2021v2 (GEOTRACES Intermediate Data Product Group, 2023). The data show ranges from 0.01 to 420 nmol kg^{-1}, with the most elevated values associated with Fe sources (anoxic Black Sea waters and/or Mid-Atlantic Ridge [MAR] hydrothermal plumes), and most data falling between 0 and 1 nmol kg^{-1}. Note nonlinear x axis on left-hand panel.

THE GEOTRACES TEI DATA "EXPLOSION"

In 2024, at the time of writing, the current GEOTRACES Intermediate Data Product 2021v2 contains over 16,000 observations of dissolved Fe (Figure 1), corresponding to about three orders of magnitude more than the pre-GEOTRACES era (Schlitzer and Mieruch-Schnülle, 2024, in this issue). This represents a veritable "explosion" in both data quality and quantity, and provides detailed, basin-scale mapping of all the oceans for many TEIs (e.g., Figure 2 for dFe). The amount of data for dFe will also increase further with the availability of the next Intermediate Data Product (IDP) in 2025. The GEOTRACES data product includes TEI data from section cruises, numerous "process" studies, and also compliant (i.e., non-GEOTRACES) datasets. It relies on rigorous intercalibration of data for inclusion and the use of "crossover stations" between sections (see Aguilar-Islas, 2024, in this issue), meaning that all data can be synthesized for comparison and interpretation and can be used to create elegant World Ocean Circulation Experiment (WOCE)-style two- and three-dimensional visualizations as in Figures 2–4 (Schlitzer et al., 2018). Such impressive cooperative production of freely available intercalibrated data and visualizations (available as an electronic atlas) is perhaps one of the greatest successes of GEOTRACES to date, ranking alongside a plethora of science outcomes (Anderson et al., 2020) and the proliferation and standardization of "clean techniques" across 35+ countries, all built on the pioneering clean collection techniques of earlier researchers (e.g., Bruland et al., 1979; Measures et al., 2008; Cutter and Bruland, 2012).

APPLICATION OF COMBINED TOOLS, EXISTING TOOLS, AND NEW TOOLS

It was clear from the very beginning of planning for a GEOTRACES-style program that a multi-tracer approach was needed to address complex marine biogeochemical questions (Jeandel, 2024,

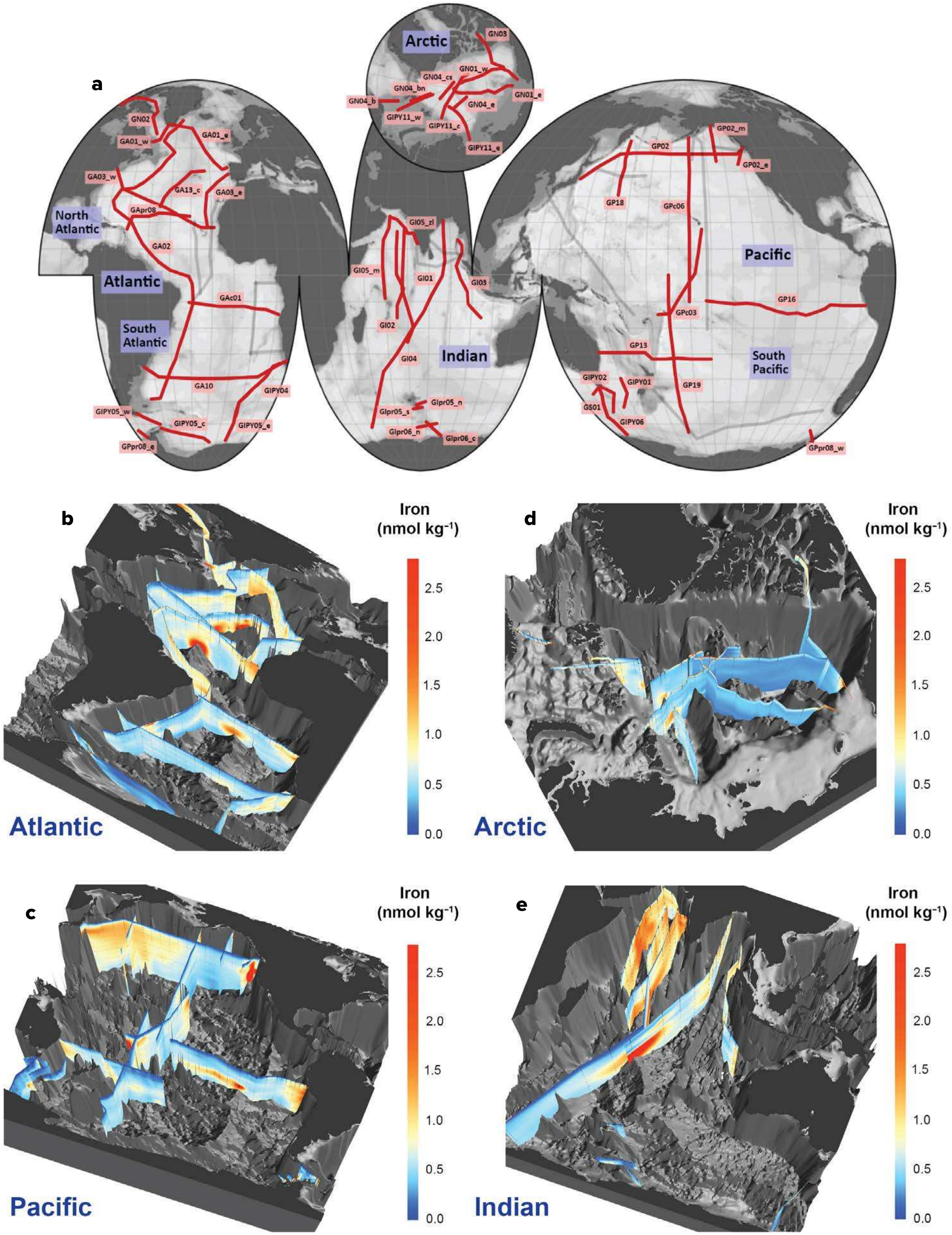

FIGURE 2. The GEOTRACES data "explosion"—a synthesis of dissolved Fe concentrations from four ocean basins. (a) A global map shows GEOTRACES cruise sections and process studies from which dissolved Fe concentrations are available in the Intermediate Data Product 2021v2 (GEOTRACES Intermediate Data Product Group, 2023), taken from the eGEOTRACES electronic atlas (Schlitzer, 2021). (b–e) Three-dimensional ocean basin views show plots of dissolved Fe concentration sections (nmol kg^{-1}) from the Atlantic, Arctic, Pacific, and Indian Oceans, respectively. *Graphics by Reiner Schlitzer, Alfred Wegener Institute*

in this issue; Anderson et al., 2014). For example, an understanding of the sources, sinks, and behavior of the bioactive elements such as Fe could be greatly enhanced by measuring the suite of micronutrient TEIs (e.g., Cd, Cu, Co, Mn, Ni, Zn) or by measuring flux (e.g., Ra, Th), particle-scavenging (e.g., Th, Pa, U, Po), boundary source addition (Al, Pb, Ra, Th, He), or circulation tracers (e.g., REE, CFCs). Accordingly, a key strength of the GEOTRACES approach and science plan, in addition to high-resolution intercalibrated datasets, is both measurement of multiple TEIs and application of these measurements to biogeochemical marine science questions (GEOTRACES Planning Group 2006; Anderson et al., 2014). Cooperative work by multiple PIs on the same ship, with samples collected from the same trace-metal clean rosette (or other coordinated sampling systems), means that a suite of different TEIs is measured on subsamples of water, particle, or aerosol samples. This is a mammoth undertaking—as an example, the recent US GEOTRACES GP17-OCE cruise in the South Pacific collected ~12,000 filtered samples for 21 laboratories (Halbeisen, 2024, in this issue)—but the benefit for understanding sources and biogeochemical processes cannot be overstated as compared to the single-TEI studies that were much more typical prior to GEOTRACES.

GEOTRACES has also stimulated the development and/or widespread application of sampling systems, techniques, and parameters. An excellent example of a "new" parameter is the application of dissolved stable Fe isotope ratios (δ^{56}Fe) to provide key insights into the sources, sinks, and internal cycling of dFe (de Jong et al., 2007; Fitzsimmons and Conway, 2023). These ratios were measured in seawater for the first time by GEOTRACES participants, with intercalibration facilitated by community exercises (Boyle et al., 2012) and subsequently applied at high resolution on at least 10 GEOTRACES sections. Other "Fe" parameters such as measurement of Fe-binding organic ligands, Fe speciation (Fe^{2+} versus Fe^{3+}), dFe size partitioning (subdivision of the 0.2 micron dFe pool into smaller operationally defined dissolved size fractions), and various forms of particulate Fe, which had all been measured to a very limited extent previously, could now be applied together at high-resolution at the basin scale (Figure 3).

The second US GEOTRACES cruise in 2011 across the subtropical North Atlantic (GA03_w) provides a particularly relevant example of all these aspects of GEOTRACES, with sections examining multiple parameters illuminating different aspects of the Fe cycle (Figure 3). For instance, dissolved Fe concentrations and δ^{56}Fe identify multiple external point sources of Fe to the section (Mid-Atlantic Ridge venting and margin sediments) set against a pervasive surface dust source across the basin, while particulate Fe highlights deep benthic nepheloid layers. Further insight into the form and longevity of Fe may be gleaned from the redox speciation, the size partitioning of the dissolved Fe (percentage colloids), and the presence of Fe binding ligands (Figure 3). Thorium isotopes (^{230}Th, ^{232}Th, ^{234}Th) have perhaps been especially useful here in the "dusty" North Atlantic, because they can provide constraints not only on dust and particle (and therefore multiple TEI) flux rates but also on export of Fe from the surface ocean as well as for residence times of multiple TEIs across the basin (see Hayes, 2024, in this issue). Radium provides further unique insights because its multiple isotopes with varying half-lives allow for TEI flux calculations, and coupling (or decoupling) of ^{228}Ra, ^{232}Th, dFe, and δ^{56}Fe in ocean sections can be used to discriminate different sources (Charette et al., 2015, 2016). For example, diffusive sediment fluxes of dFe^{2+} in the deep eastern portion of GA03_w may be indicated by benthic ^{228}Ra and low δ^{56}Fe, while the presence of near-crustal δ^{56}Fe, elevated ^{232}Th, and a lack of ^{228}Ra at intermediate depths on both margins (including within low-oxygen waters) points to lithogenic particulate fluxes and perhaps a supply of small colloidal-size lithogenic particles that would be classified as dFe rather than a large diffusive supply of reduced Fe^{2+} (Conway and John, 2014; Charette et al., 2015; Fitzsimmons et al., 2015; Hayes et al., 2018; Figure 3).

A MULTI-SOURCE VIEW OF DISSOLVED Fe

The latest combined oceanic dFe dataset (GEOTRACES Intermediate Data Product Group, 2023), which spans concentrations from just 9 pmol kg^{-1} (10^{-12} mol kg^{-1}) in remote surface waters to ~400 nmol kg^{-1} in the anoxic subsurface of the Black Sea (Figure 1, measured on Dutch GA04-N), confirms that most open oceanic dFe data are in the 0.1–1 nmol kg^{-1} range, and that lowest concentrations are found in remote surface waters that receive little dust. However, the dataset also clearly shows that the deep ocean cannot simply be characterized by a near-constant dFe concentration but instead ranges from ~0.2 to well in excess of 1 nmol kg^{-1}. Elevated dFe concentrations are found near boundary sources (dust, sediments, hydrothermal vents) and also in some cases at remarkably long distances (thousands of kilometers) from the nearest implicated boundary source (Figures 1 and 2). Such elevated dFe concentrations are thought to be not only facilitated by the presence of Fe-binding organic ligands but also dependent on the physical Fe speciation, notably the presence of colloids and via dissolved-particulate exchange (e.g., Resing et al., 2015; Fitzsimmons et al., 2017; Kondo et al., 2021; Wong et al., 2022).

So, what does the global compilation tell us about dFe sources in the different ocean basins? The major finding was the prevalence of intermediate-deep Fe sources such as hydrothermal venting or marine sediment release throughout all ocean basins (Figure 2). Indeed, it is now a remarkably safe bet that crossing a location of known high-temperature hydrothermal venting will mean observations of a dFe plume associated with that vent. Our view of the ocean must therefore now reflect multiple sources as being influential in setting the distribution of dFe (see Figure 4), while the question of how influential such sources are for driving surface productivity requires understanding of both the longevity of dFe species during transport and the regional to basin wide circulation. Although the locations of different cruise sections in different basins were laid out during GEOTRACES planning to target different and specific biogeochemical processes and boundary sources (Anderson et al., 2014), the compilation shows that the importance of different sources varies regionally, basin-by-basin (Figure 2).

A second critical aspect of the GEOTRACES dFe cycling paradigm is that dFe can be transported over

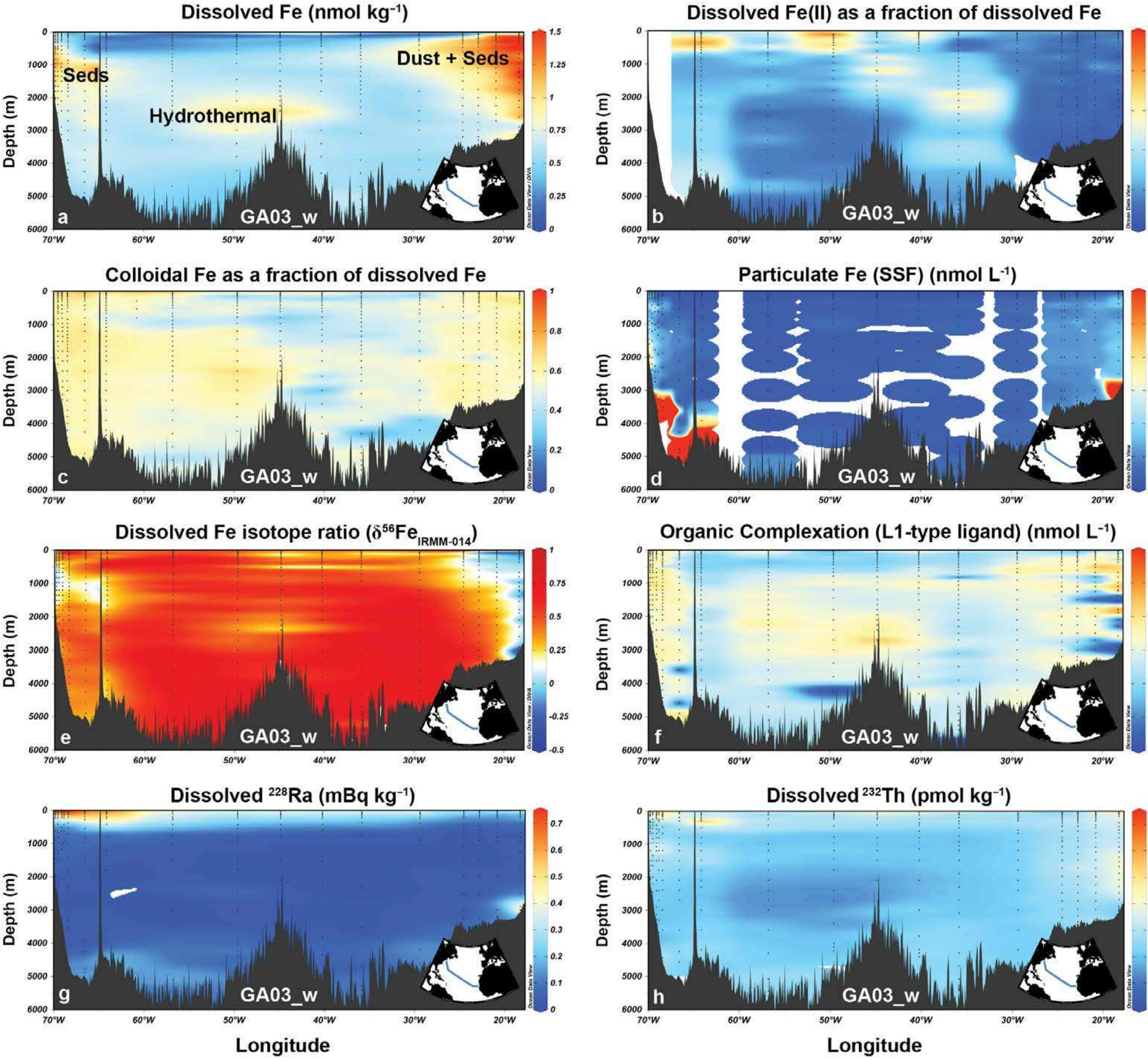

FIGURE 3. A GEOTRACES high-resolution, multiparameter approach for interrogating Fe cycling, with distributions from the subtropical North Atlantic (GEOTRACES Section GA03_w). GEOTRACES cruises sample a range of different dissolved and particulate parameters at high spatial resolution to provide a synthetic view of the processes that control elemental cycling, with an example here from the first US GEOTRACES section: (a) dissolved Fe concentration (0.2 µm size), (b) chemical speciation (fraction Fe(II)), (c) physicochemical Fe speciation (fraction colloidal Fe; ~0.02–0.2 µm size), (d) small size fraction (SSF) particulate Fe (0.8–51 µm), (e) dissolved Fe isotope ratio ($\delta^{56}Fe_{IRMM-014}$), (f) organic complexation (L1-type Fe-binding ligand concentrations), (g) dissolved ^{228}Ra (sediment diffusive flux tracer), and (f) dissolved ^{232}Th (lithogenic tracer). Data from the proximal Trans-Atlantic Geotraverse (TAG) hydrothermal plume (Mid-Atlantic Ridge crest near 26°N) sampled during the cruise are not included in sections. These data are reproduced from the GEOTRACES Intermediate Data Product 2021v2 (GEOTRACES Intermediate Data Product Group, 2023; Conway and John, 2014; Buck et al., 2015; Charette et al., 2015; Fitzsimmons et al., 2015; Hatta et al., 2015; Ohnemus and Lam, 2015; Sedwick et al., 2015; Hayes et al., 2018), and plotted using Ocean Data View (Schlitzer et al., 2023). For more geographic context of cruise locations, see Figure 2.

unexpected distances through the ocean. Several selected GEOTRACES sections in Figure 5 highlight the long-distance transport of dFe from sources such as sediments or hydrothermal vents, despite expectations that most Fe would be lost near-source, constituting a second critical aspect of the updated view in Figure 4. In fact, deep sources and transport appear to be pervasive, although intriguingly, long-distance transport is not always observed. For rivers, where most dFe was thought to be lost within estuaries during flocculation (Boyle et al., 1977), German GEOTRACES GA08 (Figure 5a) showed a dramatic and persistent plume of dFe from the Congo River for ~1,000 km into the South Atlantic, where it is thought to relieve Fe limitation (Vieira et al., 2020). Such behavior is likely to vary from river to river, as such large-scale transport was not observed for the Amazon River (Rijkenberg et al., 2014; Figure 5b). Similarly, data from German and US GEOTRACES cruises in the Arctic indicate that the transpolar drift carries riverine and shelf dFe long distances (Figure 5b; Charrette et al., 2020). Turning to the Pacific, Japanese GP02 shows a remarkable plume of sediment-derived dFe being transported across the North Pacific, up to 4,000 km away from the source in marginal seas near Japan (Nishioka et al., 2020). The mechanisms for such long-distance travel of sediment-derived dFe remain unclear, with organic-complexation invoked in most cases, while dFe may be stabilized and transported through low dissolved oxygen (Kondo et al., 2021; Wong et al., 2022). Lastly, the zonal US GP16 section across the South Pacific shows a remarkably persistent dFe plume that travels ~4,000 km from the East Pacific Rise (Resing et al., 2015; Figure 5d). This observation, although somewhat in contrast with vents in the Atlantic, reinforces the "leaky vent hypothesis" of hydrothermal venting, where a small fraction of dFe from high-Fe vent fluids, stabilized by organic-ligands or as microparticles, or from particulate-dissolved exchange, can persist over great distances throughout the ocean (Toner et al., 2012; Fitzsimmons et al., 2017; Fitzsimmons and Steffen, 2024, in this issue).

Finally, the GEOTRACES sections clearly highlight interoceanic differences in dFe distributions. For example, dust adds Fe to subtropical Atlantic surface waters, most notably near the Sahara, and there are sediment and riverine sources along the margins, as well as "bullseyes" of elevated Fe around the Mid-Atlantic Ridge (Figure 2b). However, the dominant Atlantic meridional circulation means that there is little prospect of dFe plumes spreading out zonally from sources. Further, little evidence of long-distance transport is seen in the GA02 Atlantic meridional section (Rijkenberg et al., 2014). By contrast, the Pacific, which remains less well sampled than the Atlantic, has comparatively lower dust fluxes to surface waters but notable large deep sources of Fe (sediments and hydrothermal) that travel zonally through the subsurface ocean over thousands of kilometers, facilitated by ocean circulation (Figures 2c and 5c–d). The degree to which these deep Fe sources may influence surface waters then depends on the depth of the source, the longevity of this dFe, and relevant ocean circulation, with some studies, for example, suggesting that upwelling

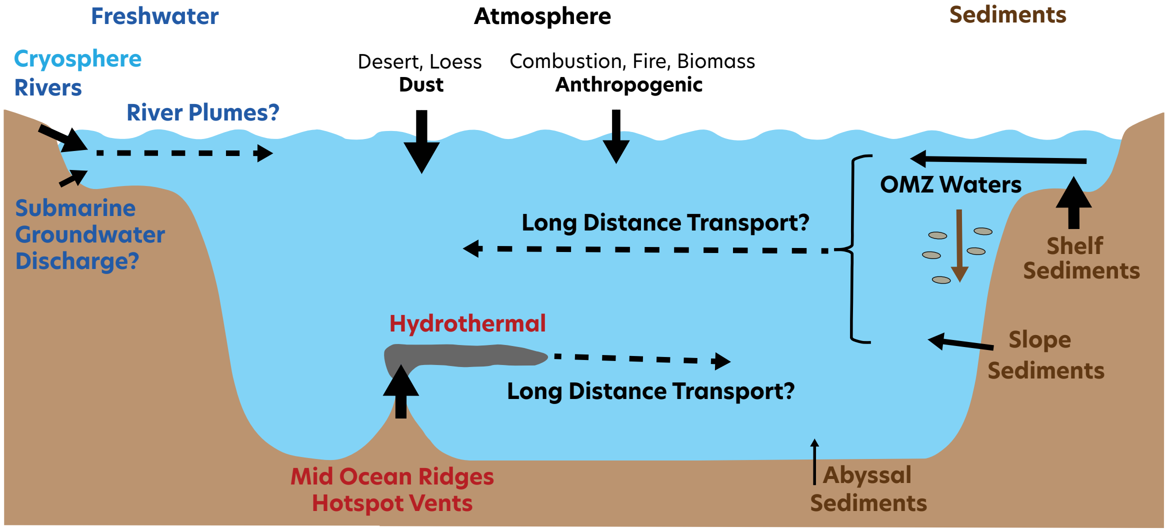

FIGURE 4. A GEOTRACES-updated multiple boundary source perspective for dissolved iron distributions in the ocean illustrates how dust, sediments, hydrothermal vents, and freshwater sources (with possible long-distance transport of Fe) all play regionally variable roles in determining marine Fe distributions. The illustration is based on the boundary source portion of the GEOTRACES Science Plan schematic (GEOTRACES Planning Group, 2006), as adapted by Conway and Middag (2024). Arrow sizes provide a representative (but inexact) view of the importance of these different fluxes, though we note this varies with ocean basin and setting, moderated by internal biogeochemical cycling processes. It is also important to note that the internal cycling processes that moderate dFe distributions are deliberately not shown (for a comprehensive view of those, see Tagliabue et al., 2017). OMZ = Oxygen minimum zone.

waters (or shallow vents) bring deeper hydrothermal Fe to the surface Fe-limited Southern Ocean (Tagliabue and Resing, 2016; Ardyna et al., 2019).

SPOTLIGHT ON MARINE SEDIMENTS AS AN Fe SOURCE

Because aerosols and hydrothermal vents are dealt with in detail by others in this special issue of *Oceanography* (Buck et al., 2024; Fitzsimmons and Steffen, 2024), here we highlight some details of marine sediments as Fe sources. Although dust was at first considered to be the primary source of Fe to the open ocean, a growing number of studies also hinted at the rival importance of marine sediments as dFe sources. Starting in the late 1990s, it was shown that sediments were an important source of dFe to surface waters along the California margin, with offshore stations also indicating that sediment-derived Fe was transported offshore at intermediate depths through lower-oxygen waters into the North Pacific (Johnson et al., 1997, 1999). Soon after, it was postulated that sediment-margin Fe fluxes—that could be transported hundreds of kilometers offshore—in fact rivaled aerosols as an ocean Fe source (Elrod et al.,

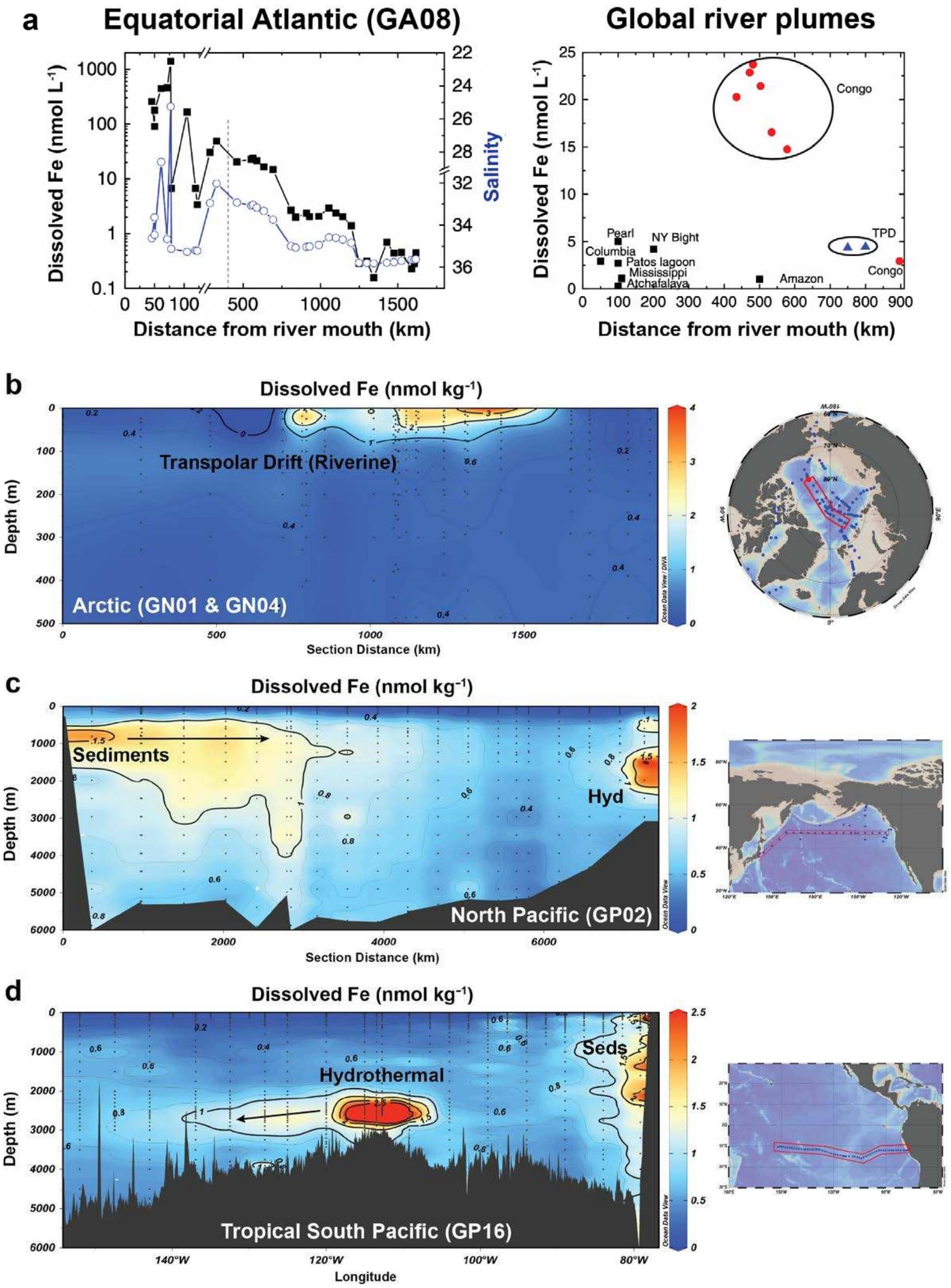

FIGURE 5. GEOTRACES datasets from multiple international groups illuminate the (non-atmospheric) oceanic boundaries as influential Fe sources to the open ocean, sometimes with subsequent long-distance transport of dFe: rivers, hydrothermal vents, and margin sediments. (a) (left) Riverine addition of Fe to the equatorial Atlantic via the Congo River Plume, and (right) the Arctic Transpolar Drift (TPD) and Congo River Plume in the context of global rivers (modified from Vieira et al., 2020). (b) Riverine and shelf additions of Fe to the Transpolar Drift in the Arctic (US-GN01 and German-GN04). (c) Long-distance transport of sediment-derived Fe from Japan to the North Pacific (Japanese-GP02). Hyd = Hydrothermal. (d) Long-distance transport of hydrothermal- and sediment-derived Fe in the South Pacific (US-GP16). Data for b–e are reproduced from the GEOTRACES Intermediate Data Product 2021v2 (GEOTRACES Intermediate Data Product Group, 2023; Resing et al., 2015; Fitzsimmons et al., 2017; John et al., 2018; Charette et al., 2020; Jensen et al., 2020; Nishioka et al., 2020; Gerringa et al., 2021), and were plotted using Ocean Data View (Schlitzer, 2023). For more geographic context of cruise locations, see Figure 2.

2004). Subsequently, and prior to the GEOTRACES field campaign, several studies indicated that sediment-derived Fe could be supplied to surface waters, especially important around islands in the Fe-limited Southern Ocean (Blain et al., 2007; Pollard et al., 2007). At the time of writing, ocean section studies of dFe have firmly established benthic sediments as an important Fe source to the ocean, possibly even the dominant Fe source to some regions via upwelling of deep waters (Tagliabue et al., 2014). Examples of long-distance transport of sediment-derived dFe away from its source has also now been demonstrated across all ocean basins (e.g., Noble et al., 2012; Conway and John, 2014; Klunder et al., 2014; John et al., 2018; Moffett and German, 2020; Nishioka et al., 2020; Jensen et al., 2020; Jensen and Colombo, 2024, in this issue). But what of the mechanisms of release and transport of this dFe, and how has GEOTRACES informed these aspects?

The classic pathway for release of dFe from sediments to the overlying water column (so-called reductive dissolution, or RD) is via diffusion of Fe^{2+} from sediment porewaters, where dFe can be present at orders of magnitude higher concentrations (micromolar levels) than in bottom waters, produced via microbial respiration of organic carbon using Fe(III) as an electron accepter (Elrod et al., 2004; Homoky et al., 2009; Severmann et al., 2010, and references therein). Here, Fe^{2+} fluxes to bottom waters are thought to be primarily controlled by organic carbon oxidation rates and bottom water oxygen conditions (Dale et al., 2015). Sediment porewater studies have been instrumental in demonstrating that the porewater Fe^{2+} reservoir has an extremely fractionated Fe isotope signature (–1‰ to –5‰) relative to marine sediments, at +0.1‰, providing a potential diagnostic tracer for sediment-derived Fe (Homoky et al., 2009; Severmann et al., 2010; Klar et al., 2017; Fitzsimmons and Conway, 2023, and references therein). Although the degree to which such low δ^{56}Fe may be attenuated during oxidative loss of Fe at the sediment-water interface or within marine bottom waters remains a sharp focus of Fe isotope research, both benthic lander and water column studies demonstrate that, under the right conditions, this distinctive light isotope signature can be transferred to elevated dFe in bottom waters, where it may persist—and even be transported over thousands of kilometers after being released into low-oxygen waters of the ocean (Severmann et al., 2010; John et al., 2018; Hunt et al., 2022). A perhaps extreme example is the transport of large fluxes of dFe from sediments into the anoxic waters of the Black Sea, where dFe concentrations as high as 400 nmol kg^{-1} have been observed (Rolison et al., 2018; Figure 1).

In addition to confirming the classic pathway for release of dFe from sediments, GEOTRACES dFe isotope studies have also been instrumental in a proposed new mechanism for dFe release from marine sediments, termed non-reductive dissolution (NRD; Radic et al., 2011). This proposed second mechanism consists of the release of lithogenic Fe(III) colloids produced by weathering (Homoky et al., 2021). Accordingly, dFe release via NRD would be decoupled from organic carbon supply and benthic oxygen conditions, and instead linked to regions of high benthic energy, sediment disturbance, and benthic nepheloid layers—thus, it would be influential in deep slope and benthic sediment environments (Homoky et al., 2021). Further, this mechanism is thought to dominate sedimentary dFe release in the deeper ocean and, unlike reductive dissolution, to release dFe with a near-crustal (+0.1‰) Fe isotope signature (Homoky et al., 2021).

Surprises from GEOTRACES include not only the prevalence of shallow, sediment-derived Fe plumes associated with subsurface oxygen minimum waters in shelf settings but also the presence of Fe plumes at deeper depths on the continental slope. There may be multiple reasons for these observations, including the non-reductive dissolution mechanism. Indeed, sediment addition at intermediate depths in the North Atlantic have been attributed to non-reductive dissolution on—or along—the oxic North American margin, or exchange with particles in deep benthic nepheloid layers (Conway and John, 2014). Turning to the productive Peru margin in the South Pacific, as perhaps expected, high fluxes of Fe^{2+} to low-oxygen bottom waters over the Peru shelf lead to elevated dFe in bottom waters and a plume of dFe(II) that is transported more than 1,000 km offshore at depths of 100–500 m within the low-oxygen core of the oxygen minimum zone, as seen on GP16 (John et al., 2018). However, work on GP16 also observed a second, unexpected, and more persistent sediment-derived dFe(III) plume emanating from the Peru margin slope under oxygenated conditions at ~1,000–3,000 m depth where reductive benthic dFe fluxes should be low (Dale et al., 2015)—a finding that defied conventional understanding and models. Possible explanations for the persistence of a deep plume included elevated flux of stabilized dFe on the slope (perhaps from NRD, sediment resuspension, or ligand binding) or re-release of dFe on the slope from Fe-rich particles sinking from the shallow plume above (John et al., 2018), the so-called "shelf to basin" shuttle of reactive Fe oxides to slope sediments (Moffett and German, 2020). Writing later, Lam et al. (2020) concluded that slope sediments may be an especially persistent source of dFe to deeper ocean waters, relevant for many margins. Each ocean margin studied adds new insight to the picture, highlighting both the excitement and the utility of these new datasets as well as the added complexity that must be considered when parameterizing global models. So far, Fe isotopes have been used to constrain either RD or NRD sediment fluxes to the water column, but it is likely that the two mechanisms of release may need to be considered together as contributors to sediment dFe release (and influencing of δ^{56}Fe) in overlying shelf and slope environments (Tian et al., 2023).

WHERE NEXT?

GEOTRACES has been wildly successful in driving forward our knowledge of the distributions of TEIs and the processes that control them. An established multi-source view of the Fe cycle can now be incorporated into models (**Figure 4**). Our understanding of the long-distance transport of Fe is strongly linked to ocean circulation. But have we ironed out the details of the mechanisms by which dFe enters the ocean? By design, GEOTRACES sections have focused on seawater collection of sufficient volumes to host multi-tracer analyses using the same water samples, limiting time available for complementary sediment coring or repeat spatiotemporal sampling. While GEOTRACES has shattered the prevailing paradigm and provided a tantalizing taste of the processes occurring near sediments or at other ocean interfaces that facilitate and/or hinder dFe release, these data have elicited many new and exciting questions regarding marine Fe cycling. Going forward, as envisaged by the GEOTRACES Science Plan, these knowledge gaps must be addressed by smaller-scale process studies based on the multi-tracer and rigorous approach of the GEOTRACES section cruises. Time series that investigate temporal change at dynamic ocean boundaries are also critical.

Taking sediments as an example, process studies need to link porewater and sediment core sampling with benthic lander rate measurements and high-resolution benthic water column measurements of multiple dissolved and particulate TEIs (including radionuclides), which could be nested in regional models of physical circulation. This would allow for a more complete understanding of the processes that facilitate TEI release and the ultimate speciation and fate of TEIs that are transported away from their sources. Development of new sampling systems for the benthic boundary layer, the shelves, and marginal seas are likely of key importance. In fact, such endeavors are already ongoing; to date, there have been ~50 GEOTRACES process studies, with a couple that have focused primarily on benthic exchange. One example is GApr04, which looked at seasonal cycling and fluxes of TEIs in the Celtic Sea (Birchill et al., 2017; Klar et al., 2017), and another is the ongoing GApr18 STING process study, which couples dFe with Ra isotopes and focuses on how submarine groundwater discharge may be the main source of Fe and dissolved organic nitrogen to the eastern Gulf of Mexico (Knapp et al., 2024). Lastly, anoxic, high-Fe sediment environments have received the most attention in studies of sediment dFe fluxes, but it is also of vital importance to understand the release of dFe in environments where Fe fluxes are low but the speciation of Fe may allow persistence and transport away from sources. Similar process studies are needed for other Fe sources such as submarine groundwater, estuaries, cryospheric settings, dust deposition, and hydrothermal venting to fully understand and constrain global fluxes of climate—and productivity-relevant dFe.

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