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An International Study of the Marine Biogeochemical Cycles of Trace Elements and their Isotopes

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ntific Committee on Ocean

GEOTRACES Science Highlights

Document prepared by: GEOTRACES IPO

Ever wonder how long your favourite element remains in the ocean before it's gone again?

- Replacement time can be quantified using large synthesised GEOTRACES datasets from the North Atlantic which can precisely define the inventory of trace elements as well as their supply rate using radioactive tracers
- Example: Ocean replacement for iron is only 6 years: it may be cycling much more quickly than previously suggested



Hayes, C. T., Anderson, R. F., Cheng, H., Conway, T. M., Edwards, R. L., Fleisher, M. Q., Ho, P., Huang, K.-F., John, S., Landing, W.M., Little, S. H. Lu, Y., Morton, P. L., Moran, S. B., Robinson, L. F., Shelley, R. U., Shiller, A. M., Zheng, X.-Y. (2018). Replacement Times of a Spectrum of Elements in the North Atlantic Based on Thorium Supply. Global Biogeochemical Cycles, 32(9), 1294–1311. DOI: http://doi.org/10.1029/2017GB005839



Gulf stream eddies are fertilizing the Western Atlantic Ocean

- Gulf Steam eddies can provide an extra supply of iron, and nutrients to the ironstarved Western Atlantic Ocean
- Authors used satellite and ocean datasets to show that these eddies may be just as important than dust in supplying iron to this area of the ocean!



Conway, T. M., Palter, J. B., & de Souza, G. F. (2018). Gulf Stream rings as a source of iron to the North Atlantic subtropical gyre. Nature Geoscience, 1. <u>http://doi.org/10.1038/s41561-018-0162-0</u>



The circulation loop in the North Atlantic and Arctic oceans depicted by the artificial radionuclides

 Three recent articles describe the journey of the two long-lived anthropogenic radionuclides iodine-129 and uranium-236 from their sources up through the Arctic Ocean and back into the North Atlantic Ocean

> The output of these works proves the potential of using ¹²⁹I and ²³⁶U as a tool for investigations on the circulation within and exchanges between the Arctic and sub-Arctic Seas

Casacuberta, N., Christl, M., Vockenhuber, C., Wefing, A.-M., Wacker, L., Masqué, P., Synal, H.-A., Rutgers van der Loeff, M. (2018). Tracing the Three Atlantic Branches Entering the Arctic Ocean With 129I and 236U. Journal of Geophysical Research: Oceans, 123(9), 6909–6921. DOI: http://doi.org/10.1029/2018JC014168



Castrillejo, M., Casacuberta, N., Christl, M., Vockenhuber, C., Synal, H.-A., García-Ibáñez, M. I., Lherminier, P., Sarthou, G., Garcia-Orellana, J., Masqué, P. (2018). Tracing water masses with 129I and 236U in the subpolar North Atlantic along the GEOTRACES GA01 section. Biogeosciences, 15(18), 5545–5564. DOI: http://doi.org/10.5194/bg-15-5545-2018

Wefing, A.-M., Christl, M., Vockenhuber, C., van der Loeff, M. R., & Casacuberta, N. (2019). Tracing Atlantic waters using 129 I and 236 U in the Fram Strait in 2016. Journal of Geophysical Research: Oceans. DOI: http://doi.org/10.1029/2018JC014399



Large increase of inputs from the land to the Arctic Ocean

- Measurements of ²²⁸Ra in 2015 U.S. Arctic cruise revealed that surface water content of this tracer has almost doubled over the last decade
- A mass balance model for ²²⁸Ra suggest that increase is due to an intensification of shelf-derived material inputs to the central basin (Figure)
- Coastal changes, in turn, could also be delivering more nutrients, carbon, and other chemicals into the Arctic Ocean and lead to dramatic impacts on Arctic food webs and animal populations



Kipp, L. E., Charette, M. A., Moore, W. S., Henderson, P. B., & Rigor, I. G. (2018). Increased fluxes of shelfderived materials to the central Arctic Ocean. Science Advances, 4(1), eaao1302. DOI: <u>http://doi.org/10.1126/sciadv.aao1302</u>



Barium isotope measurements constrain the oceanic Ba cycle

- Ba isotopes have very systematic relationship with Ba concentrations
- Provides route to assess Ba cycle in modern ocean
- And to improve proxy for paleoproductivity



Hsieh, Y.-T., & Henderson, G. M. (2017). Barium stable isotopes in the global ocean: Tracer of Ba inputs and utilization. Earth and Planetary Science Letters, 473, 269–278. <u>http://doi.org/10.1016/j.epsl.2017.06.024</u>



Nutrient co-limitation in Angola Basin

- Multiple nutrients must be supplied to stimulate phytoplankton growth
- Seawaters were amended with nitrogen, iron, and cobalt: alone and in all possible combinations
- Adding both nitrogen and iron was needed to stimulate any significant phytoplankton growth over 1000s of kilometres of ocean
- Addition of cobalt in combination with nitrogen and iron further enhanced phytoplankton growth in a number of experiments



Browning, T.J., Achterberg, E.P., Rapp, I., Engel, A., Bertrand, E.M., Tagliabue, A. and Moore, C.M., 2017. Nutrient co-limitation at the boundary of an oceanic gyre. Nature. 551, 242– 246 doi:10.1038/nature24063.



Changing the paradigm on the oceanic iron cycle

Extensive review on the recent findings on iron (Fe) cycle in the ocean

- Full understanding of any marine ecosystem cannot neglect micronutrients
- Fe oceanic sources are multiple, and supply from continental margins extends far beyond coastal zone while Fe inputs from hydrothermal activity were observed in all the oceans
- Cycling of organic iron-complexing ligands is a crucial component of the ocean iron cycle
- Phytoplankton can exhibit substantial variations in their iron stoichiometry...

Tagliabue, A., Bowie, A. R., Boyd, P. W., Buck, K. N., Johnson, K. S., & Saito, M. A. (2017). The integral role of iron in ocean biogeochemistry. Nature, 543(7643), 51–59. DOI: <u>http://dx.doi.org/10.1038/nature21058</u>





Testament of the efficiency of environmental policies

Measured lead concentrations and isotopes along the GEOTRACES sections <u>GA02</u> and <u>GA06</u>, reveal for the first time that natural lead can be detected again in the surface water of the North Atlantic

Significant proportions of up to 30– 50% of natural Pb, derived from mineral dust, are observed in Atlantic surface waters off the Sahara

This clearly reflects the success of the global effort to reduce anthropogenic Pb emissions



Bridgestock, L., van de Flierdt, T., Rehkämper, M., Paul, M., Middag, R., Milne, A., Lohan, M.C., Baker, A.R., Chance, R., Khondoker, R., Strekopytov, S., Humphreys-Williams, E., Achterberg, E.P., Rijkenberg, M.J.A., Gerringa, L. J.A., de Baar, H. J. W. (2016). Return of naturally sourced Pb to Atlantic surface waters. Nature Communications, 7, 12921. DOI: http://dx.doi.org/10.1038/ncomms12921



Oxygen biogeochemistry exerts a strong influence on cobalt cycling

The distribution of dissolved cobalt (DCo) and labile cobalt (LCo) along US East Pacific Zonal Transect (GP16) is closely tied to the oxygen minimum zone. It also shows:

(1) elevated concentrations of LCo are generated by input from coastal sources and reduced scavenging at low oxygen; (2) atmospheric deposition and hydrothermal vents along the East Pacific Rise are contrastingly minor sources of cobalt; (3) high cobalt waters are further upwelled and advected offshore; (4) phytoplankton export returns cobalt to lowoxygen water masses underneath

These processes result in covariation of DCo with oxygen and phosphates (figure)



Hawco, N. J., Ohnemus, D. C., Resing, J. A., Twining, B. S., & Saito, M. A. (2016). A dissolved cobalt plume in the oxygen minimum zone of the eastern tropical South Pacific. Biogeosciences, 13(20), 5697–5717. DOI: <u>http://dx.doi.org/10.5194/bg-13-5697-2016</u>



The coupled zinc-silicon cycle paradox illuminated

Vance and co-workers infer that the oceanic zinc distribution is the result of the interaction between the specific uptake stoichiometry in Southern Ocean surface waters and the physical circulation through the Southern Ocean hub

This work emphasizes how the consideration of 1-D cycling only can bias the understanding of (macro and micro) nutrient behaviours, and therefore their paleo-applications, although 1-D cycling may also play an important role in Zn cycling



Vance, D., Little, S. H., de Souza, G. F., Khatiwala, S., Lohan, M. C., & Middag, R. (2017). Silicon and zinc biogeochemical cycles coupled through the Southern Ocean. Nature Geoscience. DOI: <u>http://dx.doi.org/10.1038/ngeo2890</u>

[Si] (μM) [PO₄] (μM)

0 0.5

1.5



Decoupling between dissolved zinc and silicon in the North Atlantic Ocean driven by mixing of end-members

Correlation between dissolved zinc (Zn) and silicon (Si) is relatively weak in the North Atlantic Ocean (GA03 US section). Surprisingly, they present evidence that remineralization might have an insignificant effect on the zinc distribution in this region

>> Dissolved zinc in the North Atlantic Ocean is mainly controlled by water mass mixing, although some water mass end-members exhibit deviations in the Zn-Si correlation as, for example, the Mediterranean Outflow Waters.



Roshan, S., & Wu, J. (2015). Water mass mixing: The dominant control on the zinc distribution in the North Atlantic Ocean. Global Biogeochemical Cycles, 29(7), 1060–1074. doi: <u>10.1002/2014GB005026</u>



Changing the cadmium : phosphorus paradigm?

Cd/P of particles exported from the surface ocean doubles in high-nutrient low chlorophyll (HNLC) regions

Cd/PO₄ variations in the surface ocean and deep sea depend on Cd/P of degraded particles

Using a box model, authors present evidence that past changes in HNLC conditions would change the Cd- PO_4 relationship in deep sea... which has to be considered in paleoreconstructions!



Quay, P., Cullen, J., Landing, W., & Morton, P. (2015). Processes controlling the distributions of Cd and PO_4 in the ocean. Global Biogeochemical Cycles, 29(6), 830– 841. doi: <u>10.1002/2014GB004998</u>



Large fluxes of dissolved aluminium exported from the coast to the ocean

In the Eastern China Sea, the continental shelf serves as an important source of dissolved aluminium (DAI) for the overlaying waters via resuspension of sediments and benthic fluxes

More than half of this AI is transported northward within the region enclosed by the 100 m and 200 m isobaths to the Japan Sea/East Sea. The remaining flux is transported out of the shelf across the 200 m isobath

This highlights the importance of coastal processes and subsurface cross-shelf transport as a source of dissolved trace elements to the open ocean



Ren, J.-L., Xuan, J.-L., Wang, Z.-W., Huang, D., & Zhang, J. (2015). Cross-shelf transport of terrestrial AI enhanced by the transition of northeasterly to southwesterly monsoon wind over the East China Sea. Journal of Geophysical Research: Oceans, 120(7), 5054–5073. doi:10.1002/2014JC010655



Multiple controls on the dissolved aluminium fate in the western Atlantic Ocean

- atmospheric inputs are affecting only the surface and subsurface waters;
- there is an elusive but obvious coupling between Si-containing biogenic particles and Al;
- scavenging is occurring faster than the horizontal advective transports, preventing the use of AI as quantitative water mass tracer; and
- not observed at a basin-wide scale before, suspended sediments are a significant source for dissolved AI in the deep waters.



Middag, R., van Hulten, M. M. P., Van Aken, H. M., Rijkenberg, M. J. A., Gerringa, L. J. A., Laan, P., & de Baar, H. J. W. (2015). Dissolved aluminium in the ocean conveyor of the West Atlantic Ocean: Effects of the biological cycle, scavenging, sediment resuspension and hydrography. Marine Chemistry. doi: <u>10.1016/j.marchem.2015.02.015</u>



Field Data Constrain Ocean Mercury Budget

Estimates of the total amount and spatial distribution of anthropogenic mercury in the global ocean were substantially improved

Global budgets of total mercury suggest **a tripling** of the surface water mercury content and **a ~150% increase** in the amount of mercury in thermocline waters above preindustrial levels



Lamborg, C. H., Hammerschmidt, C. R., Bowman, K. L., Swarr, G. J., Munson, K. M., Ohnemus, D. C., Lam P.J., Heimbürger L-E., Rijkenberg M., Saito, M. A. (2014). A global ocean inventory of anthropogenic mercury based on water column measurements. Nature, 512(7512), 65–68. doi:10.1038/nature13563



Seasonal Iron Supply in the Southern Ocean is Dominated by Winter Mixing

Amount of iron supplied to the surface waters of the Southern Ocean is quantified

Seasonal iron supply is dominated by winter mixing with little iron input afterwards

Biological observations suggest that following the depletion of this wintertime iron pulse, intense iron recycling sustains productivity over the subsequent spring and summer

This places important constraints on how climate models represent the iron distribution and how changes in ocean physics impact iron limitation



Tagliabue, A., Sallée, J.-B., Bowie, A. R., Lévy, M., Swart, S., & Boyd, P. W. (2014). Surface-water iron supplies in the Southern Ocean sustained by deep winter mixing. Nature Geoscience, 7(4), 314–320. doi:10.1038/ngeo2101



What Controls the Copper Isotopic Composition in Oceanic Waters?

The isotopic composition of dissolved copper (δ^{65} Cu) in surface seawater is mainly controlled by supply from rivers, the atmosphere and deep seawater

The finding contradicts previous interpretations suggesting a strong role of the biological activity in δ^{65} Cu fractionation

At depth, δ^{65} Cu values are becoming heavier with the age of deep seawater

The authors built a box-model to quantify the oceanic budgets of both Cu concentrations and δ^{65} Cu. Imbalance in this model suggests that Cu fluxes from continental shelf sediment might affect Cu distribution in the open ocean



Takano, S., Tanimizu, M., Hirata, T., & Sohrin, Y. (2014). Isotopic constraints on biogeochemical cycling of copper in the ocean. Nature Communications, 5, 5663. doi:10.1038/ncomms6663



Coupling Rare Earth Elements Concentrations, Neodymium And Radium Isotopes: A Powerful Tool to Decode Environmental Processes

For the first time, neodymium (Nd) isotopic compositions have been measured together with dissolved and colloidal Rare Earth Elements (REE) concentrations in the Amazon estuary salinity gradient

Sharp drop of REE concentrations in the low-salinity area is driven by the coagulation of colloidal material

Nd isotopic ratios allow tracing that REE are released by the lithogenic material, weathered and transported by the river to the Atlantic Ocean

The original coupling with the radium (Ra) isotopes demonstrates that these dissolution processes are occurring within three weeks in the Amazon plume



Rousseau, T. C. C., Sonke, J. E., Chmeleff, J., Beek, P. van, Souhaut, M., Boaventura, G., Seyler, P., Jeandel, C. (2015). Rapid neodymium release to marine waters from lithogenic sediments in the Amazon estuary. Nature Communications, 6, 7592. doi:10.1038/ncomms8592.



Unexpected Magnitude of the Hydrothermal Iron Inputs in the Deep Pacific

GEOTRACES data demonstrate that lateral transport of hydrothermal iron, manganese and aluminium extends up to 4000 km west of the southern East Pacific Rise, therefore crossing a significant part of the deep Pacific Ocean

The dissolved iron behaves more conservatively than expected, and **the resulting flux is more than four times what was assumed before**

Results from a coupled ocean circulation/biogeochemical model demonstrate that this hydrothermal iron input may sustain a large fraction of the Southern Ocean export production



Resing, J. A., Sedwick, P. N., German, C. R., Jenkins, W. J., Moffett, J. W., Sohst, B. M., & Tagliabue, A. (2015). Basin-scale transport of hydrothermal dissolved metals across the South Pacific Ocean. Nature, 523(7559), 200–203. doi:10.1038/nature14577



Slow-spreading ridges could be major oceanic iron contributor

A large dissolved iron- and manganese-rich plume has been detected over the slowspreading southern Mid-Atlantic Ridge

It calls into question the assumption that deep-sea hydrothermal vents along slowspreading ridges were negligible contributors to the oceanic iron inventory



Saito, Mak A., Abigail E. Noble, Alessandro Tagliabue, Tyler J. Goepfert, Carl H. Lamborg, William J. Jenkins (2013) Slow-spreading submarine ridges in the South Atlantic as a significant oceanic iron source Nature Geoscience 6 (9), 775-770 DOI: <u>10.1038/ngeo1893</u>



Field data allow constraining total mercury budget

Estimates of the total amount and spatial distribution of anthropogenic mercury in the global ocean were substantially improved

Global budgets of total mercury suggest that there has been a tripling of the surface water mercury content and a \sim 150% increase in the amount of mercury in thermocline waters



Lamborg, C. H., Hammerschmidt, C. R., Bowman, K. L., Swarr, G. J., Munson, K. M., Ohnemus, D. C., Lam P.J., Heimbürger L-E., Rijkenberg M., Saito, M. A. (2014). A global ocean inventory of anthropogenic mercury based on water column measurements. Nature, 512(7512), 65–68. doi:10.1038/nature13563



Dissolved iron sources in the North Atlantic Ocean quantified

Using a novel method based on the stable isotopic composition of dissolved Fe, the relative importance of four different dissolved iron (Fe) sources in the North Atlantic Ocean have been precisely determined for the first time

Results (contribution of dissolved iron): Saharan dust: 71-87 % North American margin sediments: 10-19 % African margins contributes: 1-4 % Hydrothermal venting Mid-Atlantic Ridge: 2-6 %

Tim M. Conway, Seth G. John (2014) Quantification of dissolved iron sources to the North Atlantic Ocean, Nature, 511(7508), 212–215. doi:10.1038/nature13482.





Copper and zinc oceanic mass balance revisited

...with the original approach that takes into account the hitherto ignored constraint of their isotopes

Copper (Cu) oceanic mass balance appears to be roughly in balance, but the zinc (Zn) one is far from being constrained... isotopes reveal that either an "isotopically light sink" or "isotopically heavy source" is missing



Little, S. H., Vance, D., Walker-Brown, C., & Landing, W. M. (2014). The oceanic mass balance of copper and zinc isotopes, investigated by analysis of their inputs, and outputs to ferromanganese oxide sediments. Geochimica et Cosmochimica Acta, 125, 673–693. doi:10.1016/j.gca.2013.07.046



First simultaneous method for the determination of iron, zinc and cadmium isotopes in small volumes of seawater

Designed for use on *only a single litre* of seawater representing a 1–20 fold reduction in sample size and a 4–130 decrease in blank

The procedure yields data with high precision, allowing estimation of natural variability in the oceans, which spans 1–3‰ for all three isotope systems



Tim M. Conway, Angela D. Rosenberg, Jess F. Adkins, Seth G. John (2013), A new method for precise determination of iron, zinc and cadmium stable isotope ratios in seawater by double-spike mass spectrometry, Analytica Chimica Acta, Volume 793, Pages 44-52, DOI: 10.1016/j.aca.2013.07.025



GEOTRACES scientists discover new variability in iron supply to the oceans with climate implications

The amount of dissolved iron released into the ocean from continental margins displays variability not currently captured by oceanclimate prediction models. The amount of iron leaking from continental margins sediments was previously assumed to reflect rates of microbial activity within the sediments

Dr. William Homoky and co-authors found that rate of iron released from seafloor sediments close to continents is actually far more varied between regions because of local differences in weathering and erosion on land



Homoky, W. B. et al. Distinct iron isotopic signatures and supply from marine sediment dissolution (2013), Nature Communications, 4:2143, DOI: 10.1038/ncomms3143



A global compilation of dissolved iron measurements: focus on distributions and processes in the Southern Ocean

A data synthesis effort of over 13,000 observations of dissolved iron concentrations. More than doubled the previous data compilation

Overall, more observations have been collected in the past 5 years under the auspices of the International Polar Year and GEOTRACES efforts than were collected in the prior ~15 years

Alessandro Tagliabue's and co-authors analysis was able to highlight where observations are lacking in a particular region or time of year, which they hope will assist future sampling efforts

Tagliabue, A., et al. (2012) A global compilation of dissolved iron measurements: focus on distributions and processes in the Southern Ocean, Biogeosciences, 9, 2333-2349, doi:10.5194/bg-9-2333-2012.



Substantial intra-basin variation of the dissolved metal/phosphorus ratio in the different water masses of the Indian Ocean

The first simultaneous, full-depth, and basin-scale section-distribution of dissolved AI, Mn, Fe, Co, Ni, Cu, Zn, Cd and Pb in the Indian Ocean (GI04), show an important variability of the dissolved metal/phosphorus ratio among the water masses

These results question the validity of using an "extended Redfield ratio" to trace metals. The consistent mechanism yielding these variations remains to be understood.



Thi Dieu Vu, H., Sohrin, Y. (2013) Diverse stoichiometry of dissolved trace metals in the Indian Ocean, Scientific Reports 3, <u>DOI: 10.1038/srep01745</u>



New beautiful results on marine particle speciation, a challenge for the GEOTRACES community...

Advanced Light Source x-ray spectromicroscopy (XANES) allows a fine description of the marine Fe pool chemical speciation and mineralogy

The paper identifies a diverse array of iron particles, showing impressive variations in the oxidation state and composition of iron particles between the coasts of South Africa and Antarctica

As particle speciation is directly linked to the element solubilities, these differences may affect the production of bioavailable dissolved iron



B. P. von der Heyden, A. N. Roychoudhury, T. N. Mtshali1, T. Tyliszczak, S. C. B. Myneni. (2012). Chemically and Geographically Distinct Solid-Phase Iron Pools in the Southern Ocean: Science 338 (6111): 1199-1201.



Arsenic detoxification by phytoplankton reveals that arsenic species could be good proxies of phosphorus limitation

Some phytoplankton species have the capacity to modify surface water arsenic speciation, inhibiting its toxicity when phosphate concentrations are below those for arsenic (As)

During GEOTRACES cruise (GA03), fine determination of As speciation allowed establishing the potential use of these detoxification products as indicators of phosphorus (P) limitation



Wurl, O., L. Zimmer, and G.A. Cutter. 2013. Arsenic and phosphorus biogeochemistry in the ocean: Arsenic species as proxies for P-limitation. Limnol. Oceanogr. 58: 729-740.



Original Results in the Southern Ocean on Mercury speciation and Cadmium isotopes

For the first time detailed mercury (Hg) speciation was determined along an Australia-Antarctic section (Cossa, et al., 2011) and cadmium (Cd) isotopes along a South Africa-Antarctic section (Abouchami W., et al., 2011). Both were acquired along two IPY-GEOTRACES sections.

Hg species distribution suggests distinct features in the Southern ocean Hg cycle: (i)net atmospheric Hg deposition on surface water near the ice edge, (ii) Hg enrichment in brines during sea-ice formation and (iii) a methylmercury maximum close to Antarctica, far from anthropogenic Hg emissions (see figure below).



Cossa, D., Heimbürger, L.-E., Lannuzel, D., Rintoul, S.R., Butler, E.C.V., Bowie, A.R., Averty, B., Watson, R.J., Remenyi, T., (2011), Mercury in the Southern Ocean, Geochimica et Cosmochimica Acta, 75(14): 4037-4052 DOI: 10.1016/j.gca.2011.05.001



Original Results... (cont.)

Cd isotope ratios and concentrations are distributed into two distinct Cd isoscapes,delimiting two biogeochemical provinces the Antarctic Circumpolar Current(ACC)andWeddellGyre(seefigure).

A doubling of the isotope effect due to biological consumption of Cd is observed during water transport from the Weddell Sea into the ACC. The increase in the magnitude of Cd isotope fractionation, as a result of enhanced phytoplankton biomass and altered species composition in the Southern Ocean, demonstrates that Cd isotopes could potentially serve as a useful measure of biological productivity.

Abouchami W., Galer S.J.G., de Baar H.J.W., et al. (2011), Modulation of the Southern Ocean cadmium isotope signature by ocean circulation and primary productivity, Earth and Planetary Science Letters, 305(1-2) DOI: 10.1016/j.epsl.2011.02.044





Measuring precisely with only 40 ml of sample



It is now possible to measure simultaneously and precisely manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd), and lead (Pb) concentrations in seawater using only 40 ml of sample.

All the cooking recipes and techniques are given in the recent detailed article of Dondra Biller and Kenneth Bruland in Marine Chemistry (Biller and Bruland, 2012).

Biller Dondra V., Bruland Kenneth W. (2012) Analysis of Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb in seawater using the Nobias-chelate PA1 resin and magnetic sector inductively coupled plasma mass spectrometry (ICP-MS) Marine Chemistry 130-131 (2012) 12–20 DOI: 10.1016/j.marchem.2011.12.001

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