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Is Hg recycling into the Earth's mantle negligible?

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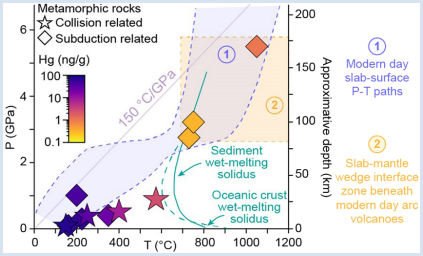
Abstract



Mercury (Hg), with its ultra-trace abundance in rocks and high volatility, challenges the investigation of its distribution and mobilisation in geological systems. Based on similar Hg isotope mass independent fractionation (*i.e.* Δ^{199}Hg) values between marine and terrestrial sediments and magmatic rocks, it is commonly suggested that Hg has been continuously exchanged between the Earth's mantle and the atmosphere through subduction recycling. However, a review of Hg concentrations in ophiolites and orogenic peridotites, mantle xenoliths, mantle derived basalts and mafic rocks (Mg# ≥60), and variably metamorphosed collision and subduction related rocks suggests otherwise. Ultra-trace Hg concentrations in mantle rocks and mantle derived magmas are inconsistent with Hg recycling into the mantle. In fact, collision and subduction related rocks progressively lose Hg with increasing metamorphic grade, indicating that only ≤1 ng/g of Hg is transferred to the sub-arc mantle melting regions. These observations also suggest that Hg recycling was inhibited during the Archean. Here we warn about the risk of using Hg isotopes alone to interpret the Hg cycle on Earth and urge the need for new and accurate Hg concentrations in crystalline rocks from different geological settings.

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Introduction

Sediments and seawater altered oceanic basalts undergo significant loss of mercury (Hg) during prograde metamorphism due to its high volatility, resulting in high grade metamorphic rocks (>500 °C and ~0.5–3.0 GPa) having Hg concentrations <3 ng/g (Marowsky and Wedepohl, 1971; Pitcairn *et al.*, 2006, 2010, 2015; Stepanov, 2021; Chen *et al.*, 2025). On a global scale, such low Hg concentrations in these rocks question the efficacy of Hg recycling into the mantle *via* subduction. To date, however, the large variation in Hg isotope mass independent fractionation values (typically as Δ^{199}Hg) between basaltic rocks (−0.6 to +0.4 ‰) and terrestrial and marine sediments (−1 to +0.4 ‰) is taken as evidence of the Hg exchange between the Earth's lithosphere and the atmosphere-land-ocean systems (*e.g.*, Yin *et al.*, 2024; Xu *et al.*, 2025 and references therein).

Such an apparent contrasting behaviour between elemental Hg and its isotopes defies whether the Hg concentrations observed in mantle rocks and mantle derived magmas reported in the literature support the recycling of Hg into the Earth's mantle. Degassing processes in magmatic and volcanic systems modulate the final Hg concentration in rocks (*e.g.*, Bouilliung *et al.*, 2025). Also, analytical artefacts linked to low instrumental reproducibility and/or sample contamination can alter the measurement precision of Hg concentration in rocks (Narduzzi *et al.*, 2025). Here, we propose an alternative scenario for the Hg geochemical cycle by reviewing Hg concentrations in mantle peridotites, mantle derived basalts and mafic rocks (Mg# [Mg/(Mg + Fe_{tot}) mol. %] ≥ 60), and rocks from variably metamorphosed collision and subduction related settings. We aim to demonstrate the necessity of establishing new constraints on Hg concentration in protoliths and its behaviour during metamorphism. Details of data filtering procedure are reported in the Supplementary Information, along with the Hg and major element concentrations of the used samples (Table S-1). Data are presented as median ± interquartile range or as median ± standard deviation (1σ) when Hg analyses in the data set were ≤4. In a few cases, only one analysis was reported.

Mercury in the Earth's Mantle and Mantle Derived Rocks

Mantle peridotites, mantle derived basalts and mafic rocks exhibit similar Hg concentrations ranging from $1.0^{+0.5}_{-0.4}$ to $2.1^{+3.0}_{-1.1}$ ng/g (Fig. 1). However, most of these data are acquired *via* thermal decomposition atomic absorption spectrometry (TD-AAS) using DMA-80 and Lumex RA 915+, which can potentially produce relatively inaccurate and imprecise results when Hg concentrations in rocks are <10 ng/g (Narduzzi *et al.*, 2025). Most of the previous studies rarely provide statistically robust results and a comprehensive description of the employed

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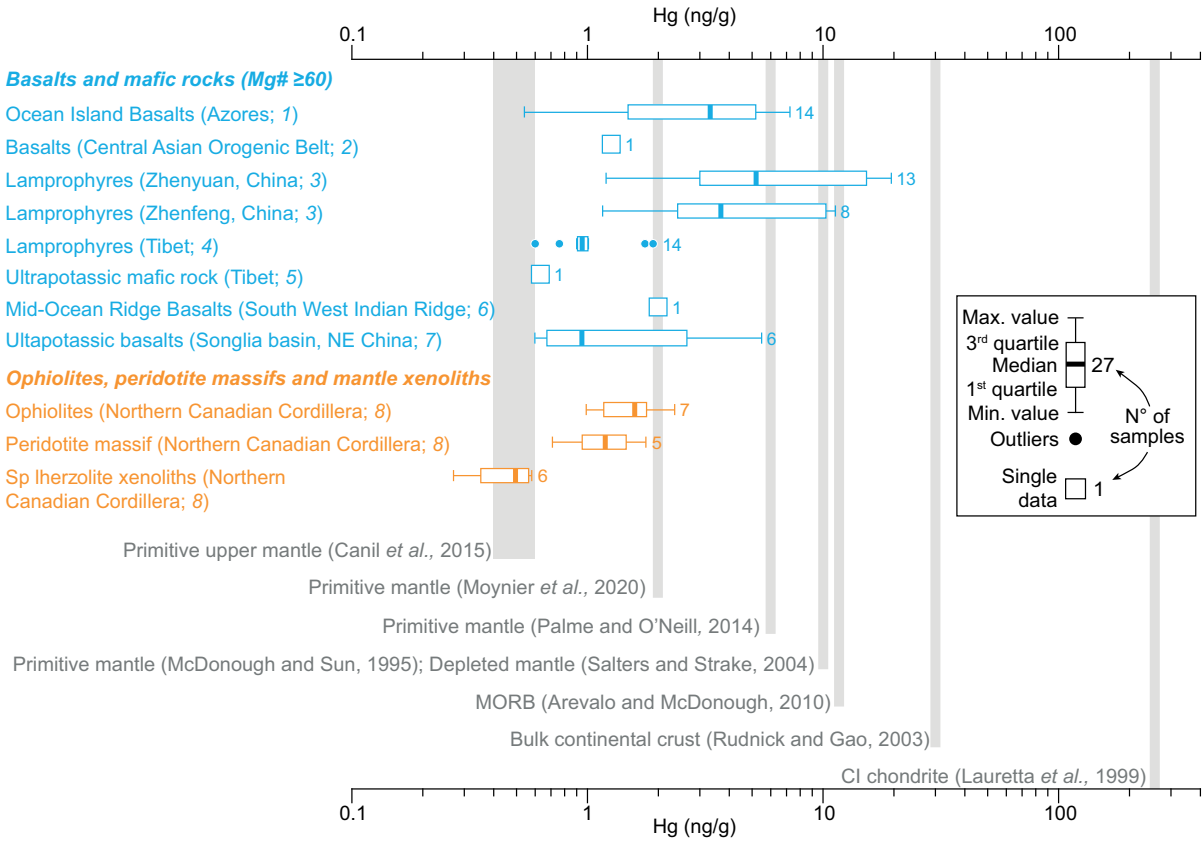

Figure 1 Box plot diagram showing the mercury (Hg) concentrations (ng/g) in mantle rocks, mantle derived basalts and mafic rocks, depleted mantle (DM), MORB, bulk continental crust, CI chondrite and primitive mantle. Numbers in parentheses correspond to references in Table S-1, which are also reported in the SI.

analytical methodology in terms of specifics regarding a single or set of analyses *per* sample, the related standard deviations, sample weights associated to the data, and procedural blanks prior to the sample measurements. This lack of information casts doubts on the uncertainty and repeatability of the results.

For instance, the Hg concentration in mid-ocean ridge basalts (MORBs) (12.5 ng/g; Arevalo and McDonough, 2010) appears to be unreliable (Fig. 1) because it was derived from Salters and Stracke's (2004) uncertain depleted mantle (DM) Hg value. Salters and Stracke (2004) suggested 10 ng/g of Hg for the DM (Fig. 1) based on the similar Hg/Mn ratio between the primitive mantle (PM) of McDonough and Sun (1995) and the bulk continental crust (CC) of Rudnick and Gao (2003). Such a Hg concentration in the DM represents only an approximation because Hg is rarely measured in basalts and peridotites, and the CC does not always complement the DM (Salters and Stracke, 2004). Additionally, the Hg value of the PM of McDonough and Sun (1995) (~10 ng/g) derives from limited flameless AAS analyses on basaltic and peridotitic reference materials (Flanagan *et al.*, 1982) and CV-AAS analyses, after dissolution with aqua regia/HF, on orogenic peridotites from the Ivrea-Verbano Zone (Italy, Garuti *et al.*, 1984), the latter being likely contaminated (Palme and O'Neil, 2014; Canil *et al.*, 2015; Narduzzi *et al.*, 2025).

The estimate of Hg in the PM (~6 ng/g) provided by Palme and O'Neill (2014) considers that Hg is chalcophile and the Hg/Se ratio, calculated using the CC of Gao *et al.* (1998), is constant during mantle melting. However, the Hg concentration in the PM might be biased because the Gao *et al.* (1998) CC Hg concentration (~9 ng/g) is based on East China rocks rather than on a global data set (*e.g.*, Rudnick and Gao, 2003). Conversely, Moynier *et al.* (2020) obtained ~2 ng/g of Hg for the PM, assuming that Hg was delivered by late accretion. However, it is unknown whether this value would change if Hg was partially or entirely partitioned into the core and/or devolatilised during planetary accretion. The work of Canil *et al.* (2015) provides an exemplary approach to carefully assessing the uncertainties with Hg concentrations in rocks and how these are related to sample preparation procedures and instrumental errors. Indeed, Canil *et al.* (2015) proposed 0.4–0.6 ng/g of Hg for the primitive upper mantle (PUM), based on TD-AAS analyses of six spinel lherzolite xenoliths from British Columbia showing depletion/enrichment trends. Besides the TD-AAS derived uncertainties (see Narduzzi *et al.*, 2025), the PUM value is based on a limited and local mantle xenolith dataset, and therefore not likely to be representative of the whole Earth's mantle.

Mercury concentrations in mantle peridotites are below ~2.5 ng/g (*i.e.* maximum value from ophiolites; Canil *et al.*, 2015). Note that here the analytical methods employed were mostly TD-AAS. As reliable results of cold vapour atomic fluorescence spectroscopy (CV-AFS) usually correspond to the minimum TD-AAS values (Narduzzi *et al.*, 2025), we suggest that the actual Hg concentrations in mantle peridotites range between 0.25 and 1.0 ng/g. This is below the Moynier *et al.* (2020) PM Hg value and partially overlaps with the Canil *et al.* (2015) Hg range in the PUM. The generally lower Hg concentrations in peridotites compared to most PM models are consistent with the incompatible behaviour of Hg during mantle melting and its progressive partitioning into the crust. Based on the above considerations on the analytical uncertainties, the Hg concentration in mantle derived magmas is most likely between ~0.5 and 2 ng/g. Such a small difference between peridotites and mantle

derived basalts would suggest relative partition coefficients for Hg (KD values for peridotite/melt are between 0.1 and 2.0) higher than previous estimates (Canil *et al.*, 2015). Alternatively, this might reflect the use of Hg concentrations from magmatic rocks that underwent extensive Hg loss due to degassing processes before complete solidification. This would be consistent with the experimental observations of Bouilliung *et al.* (2025), who reported 99 % of Hg degassing from basaltic melts at atmospheric pressure.

Mercury in Metamorphic Terrains

Here, we report the metamorphic terrains in which Hg analyses were obtained for samples that underwent different peak metamorphic conditions and along different geothermal gradients (Table S-1 and Fig. S-1). Unless otherwise discussed, the studies summarised below used samples from areas where post-metamorphic magmatism has not occurred and represent prograde metamorphic rocks.

The Otago and Alpine Schists, New Zealand. These terrains form a <180 Ma accretionary complex where unmetamorphosed (100–200 °C, ≤0.1 GPa) sediments and seawater altered seafloor basalts underwent similar losses of Hg during prograde metamorphism up to amphibolite facies conditions (550–650 °C, 0.8–1.0 GPa; Pitcairn *et al.*, 2006, 2010, 2015). Unmetamorphosed and amphibolite facies rocks have Hg concentrations of 75^{+10}_{-13} ng/g and $2.6^{+0.3}_{-1.2}$ ng/g, respectively (CV-AFS analyses). Such data implies a potential Hg loss of ~97 % with increasing metamorphic grade (Fig. S-1a). Specifically, the greatest loss of Hg occurs at or before 300–500 °C (greenschist facies), where Hg decreased to $8.1^{+8.8}_{-4.2}$ ng/g, equivalent to ~89 % decrease of Hg concentration.

California Coastal Range, USA. Mercury was analysed (cold vapour MC-ICP-MS) in some lithologies near Clear Lake (Smith *et al.*, 2008). The metamorphic P-T peak conditions were retrieved from other works (Fig. S-1b). Sediments from Great Valley (163 ± 18 °C, ~0.1 GPa) and Franciscan Complex (225 ± 35 °C, ~0.3 GPa) share similar Hg concentrations (65^{+22}_{-10} and 64^{+21}_{-22} ng/g, respectively) but are high compared to the Franciscan blueschists (200 ± 50 °C, 1 ± 0.5 GPa) and Coastal Ranges ophiolitic serpentinites (350 °C, ~0.4 GPa), which have 43 ± 11 and 24 ± 6 ng/g of Hg, respectively.

Dabie and Sumdo Belts, China. Dabie (2.8–3.6 GPa, 700–800 °C) and Sumdo (~2.7 GPa, ~730 °C) retrogressed eclogites (Fig. S-1c,d) share similar Hg concentrations ($Hg = 0.3^{+0.4}_{-0.0}$ and $0.3^{+0.1}_{-0.1}$ ng/g, respectively) as analysed *via the* Lumex RA 915+ technique (Chen *et al.*, 2025). Such Hg concentrations are lower than their assumed protoliths, with Hg mean concentrations of 13.5 ± 8.9 ng/g and 1.36 ± 0.55 ng/g for the Dabie and Sumdo protoliths, respectively, (Chen *et al.*, 2025). Accordingly, 70 % of Hg was released from the subducting slab while at crustal depths, and <30 % of Hg entered the mantle (Chen *et al.*, 2025). However, the percentages of Hg release reported in this paper are meaningless as the protoliths are unrelated in space and/or time to the Sumdo and Dabie eclogites. The protoliths they use in this study are basalts from present day mid-oceanic ridge settings and granitoids from the northeastern Central Asian Orogenic belt and South China Craton (*cf.* Fig. 1a in Chen *et al.*, 2025), and their Hg concentrations do not reflect those of the protoliths of the studied eclogites.

Central Slave Craton. Mercury analyses (LA-ICP-MS) were also performed on sulphide inclusions in eclogite diamonds, which formed during Paleoproterozoic subduction of an oceanic crust interacting with serpentinite derived fluids (Fig. S-1e; Aulbach *et al.* 2012). According to nitrogen systematics of diamonds and a 1.85 Ga formation age for the sulphides, diamond mantle residence T was ~1050 °C, corresponding to a depth of ~170 km (~5.5 GPa; Aulbach *et al.*, 2012). Here, although >30 % of Hg analyses were below the LA-ICP-MS detection limit (0.158 ± 0.115 µg/g), Aulbach *et al.* (2012) calculated on the remaining data that the bulk eclogite xenolith had ≤1 ng/g of Hg (Fig. S-1e).

Implications and Conclusions

The estimated concentrations of Hg in mantle derived rocks (Fig. 1) cast doubts on the possibility that significant amounts of Hg are recycled into the Earth's mantle. Accordingly, the Hg concentrations in both collision and subduction related metamorphic rocks show a systematic decrease with increasing metamorphic grade (Fig. 2). Indeed, Hg devolatilisation appears to increase with temperature while being independent of pressure (Pitcairn *et al.*, 2006, 2010, 2015; Stepanov, 2021 and references therein). Consequently, by inference, we propose that any lithology along the modern day subduction P-T paths may lose Hg due to an increase in temperature (Fig. 3), and that more than 97 % of Hg can be devolatilised from any lithology forming a subducting slab before crossing the sediment and oceanic crust wet solidus curves, hence before reaching the slab-mantle wedge interface zone beneath arc volcanoes (Fig. 3; Grove *et al.*, 2012; Leeman, 2020). This observation is well supported by the Dabie and Sumdo belt eclogites which have negligible Hg (~0.3 ng/g) and plot within the P-T range conditions of modern day subduction and the slab-mantle wedge interface zones (Fig. 3). Based on these observations, and in the absence of data that proves otherwise, it can be reasonably proposed that only a negligible amount of Hg (≤1 ng/g) can be recycled into the Earth's mantle nowadays. The high solubility of Hg in crude oil (Wilhelm and Bloom 2000) indicates that methane-rich fluids and petroleum forming within the oil-gas window (~60–225 °C, ≤0.25 GPa; Stepanov, 2021; Goldfarb and Pitcairn, 2023) and their migration towards shallower levels are likely responsible for the tremendous loss of Hg (up to ~89 %) during metamorphism up to greenschist facies conditions (Stepanov, 2021; Goldfarb and Pitcairn, 2023).

High upper mantle temperatures during the Archean (~1450–1800 °C; *e.g.*, Palin *et al.*, 2020) that would have hindered oceanic lithosphere subduction (*e.g.*, Brown *et al.*, 2020; Palin *et al.*, 2020), and the observation that ~97 % of Hg is lost before oceanic crust and sediment wet-melting (Fig. 3), imply that recycling of Hg into the early Earth's mantle was also negligible. For instance, the bulk Hg calculated for the ~1.85 Ga Central Slave Craton eclogite xenolith (Aulbach *et al.*, 2012) indicates that ≤1 ng/g of Hg was recycled into the Paleoproterozoic mantle. Eventually, the limited amount of Hg recycled into the mantle over time (≤1 ng/g in the rocks of the subducted slabs), suggests that most of the Hg in present day volcanic rocks (>0.5–2.0 ng/g) might either derive from a primordial mantle reservoir, from assimilation of relatively Hg enriched crustal material, or both.

In summary, this review highlights that the behaviour of Hg in different geological conditions and settings still remains to be well constrained, a problem exacerbated by the unforeseen uncertainties of the analytical methods used to analyse this element. While Hg isotopes suggest recycling of Hg into the mantle (*e.g.*, Yin et al., 2024; Xu *et al.*, 2025), our review suggests that Hg is almost entirely devolatilised before the subducting slab reaches the P-T conditions that allow fluids to migrate towards sub-arc mantle melting regions. Because Hg recycling was likely also inhibited in the Archean and Paleoproterozoic mantle, we then expect that Hg exchange between Earth's

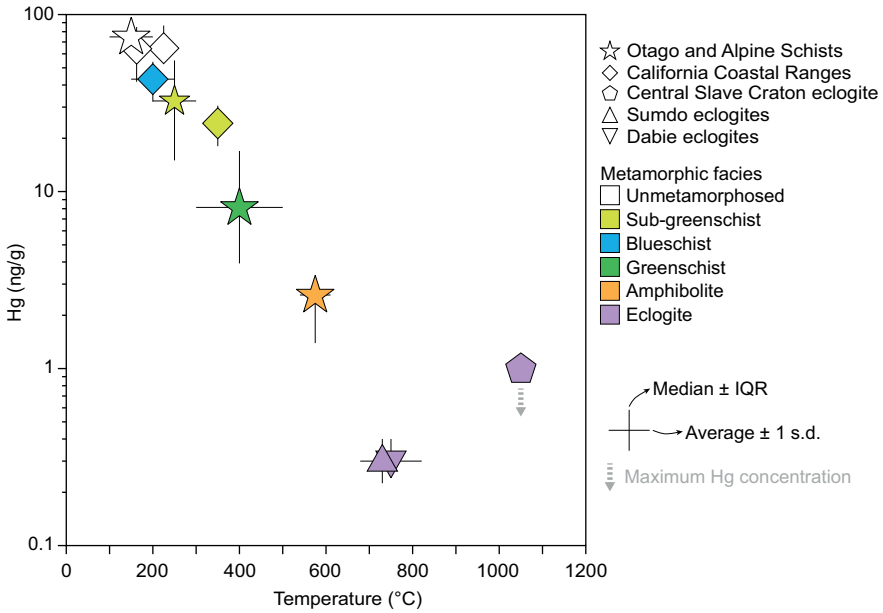

Figure 2 Mercury (Hg) concentrations (ng/g) *vs*. temperature (°C) in metamorphic samples from Otago and Alpine schists, New Zealand, California Coastal Ranges, USA, Dabie and Sumdo belts, China, and Central Slave Craton, Canada.

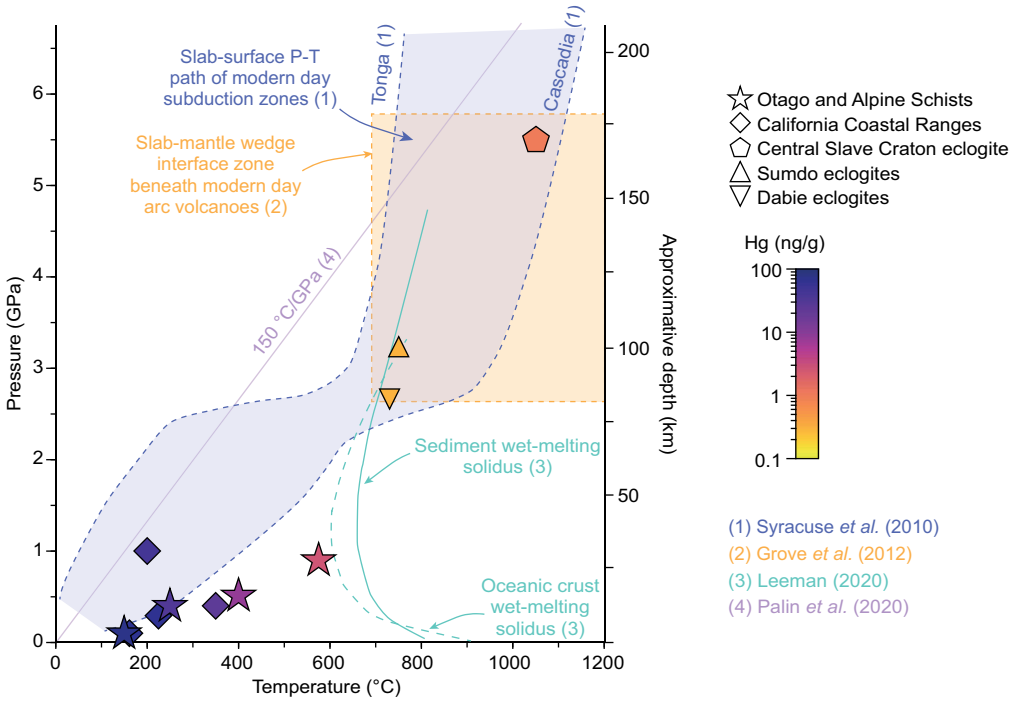

Figure 3 P-T diagram showing the devolatilisation of Hg. Mercury concentrations (ng/g) are median values discussed in the text. Tonga and Cascadia (Syracuse *et al.*, 2010) represent the coldest and hottest subduction zones (after Leeman, 2020). The geothermal gradient of 150 °C/GPa (Palin *et al.*, 2020) is reported solely for reference.

lithosphere and the atmosphere-land-ocean systems was very limited over time. Hence, it appears that Hg isotopes only track a minimal part of the Hg cycle, whereas the majority of Hg lost during devolatilisation remains unsampled. A more robust way of calculating the cycling of Hg into the mantle might be using the existing data set highlighted in this manuscript, with Hg concentrations in forearc sedimentary and metamorphic lithologies and in volcanic rocks from arc settings. We emphasise the importance of using methodologies that allow for measurements of Hg with uncertainties ≤1 ng/g, and of reporting the complete analytical procedure that allows for assessing the uncertainty of the measurements. Only with such a data set it will be possible to test the hypothesis suggested in this work and to quantitatively estimate the distribution of Hg in the subduction cycle.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2626.

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