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Is Hg recycling into the Earth's mantle negligible?

F. Narduzzi[1*], S. Covelli[2], M. Pistone[3], I. Pitcairn[4], R. Tribuzio[1,5,6], L. Ziberna[2]

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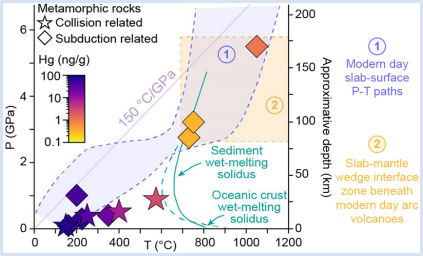
Abstract



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

1. Dipartimento di Scienze della Terra e dell'Ambiente, Università di Pavia, 27100, Pavia, Italy
2. Dipartimento di Matematica, Informatica e Geoscienze, Università degli Studi di Trieste, 34128 Trieste, Italy
3. Department of Geology, Franklin College of Arts and Sciences, University of Georgia, Athens, GA 30602-2501, USA
4. Department of Geological Sciences, Stockholm University, 10691, Stockholm, Sweden
5. Istituto di Geoscienze e Georisorse, C.N.R., Unità di Pavia, 27100 Pavia, Italy
6. Istituto Nazionale di Oceanografia e di Geofisica Sperimentale, 34010 Sgonico, Trieste, Italy
* Corresponding author (email: francesco.narduzzi@unipv.it)

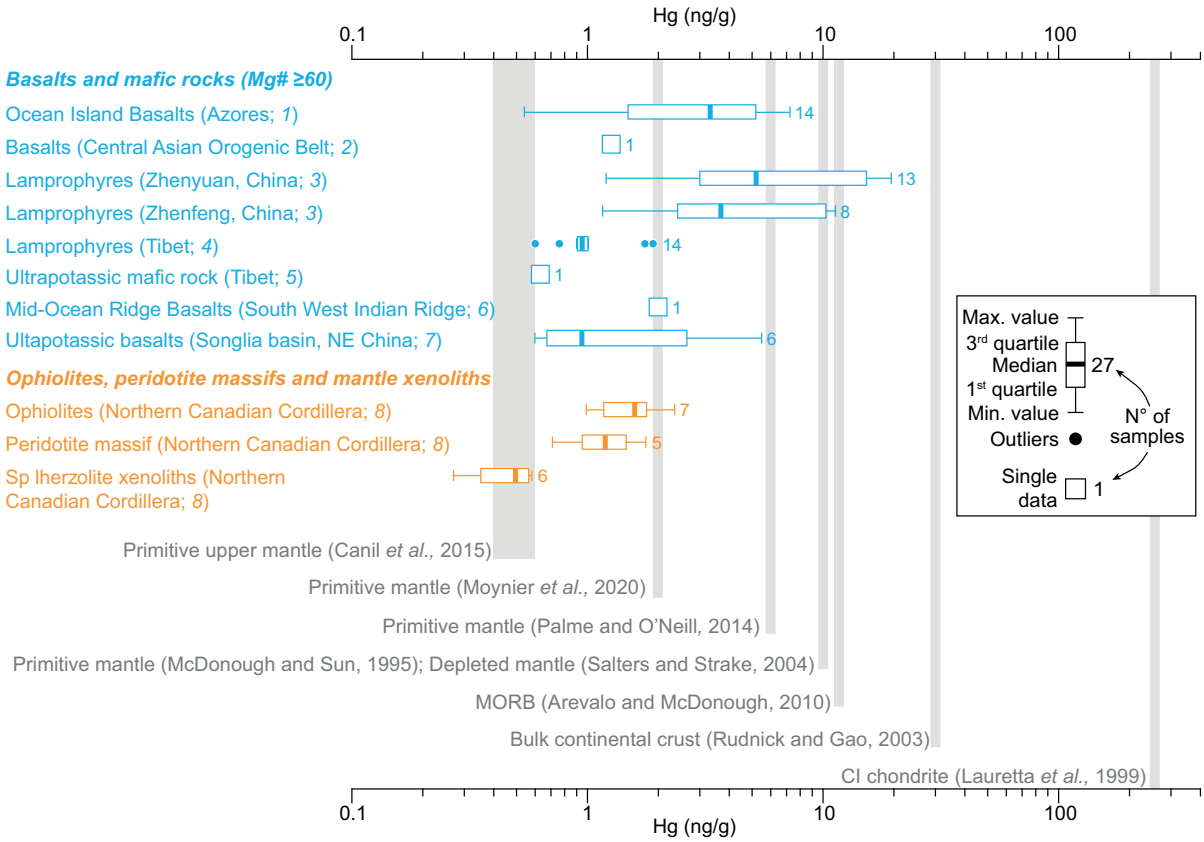

Figure 1 Box plot diagram showing the mercury (Hg) concentrations (ng/g) in mantle rocks, mantle derived basalts and mafic rocks, depleted mantle (DM), MORB, bulk continental crust, CI chondrite and primitive mantle. Numbers in parentheses correspond to references in Table S-1, which are also reported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

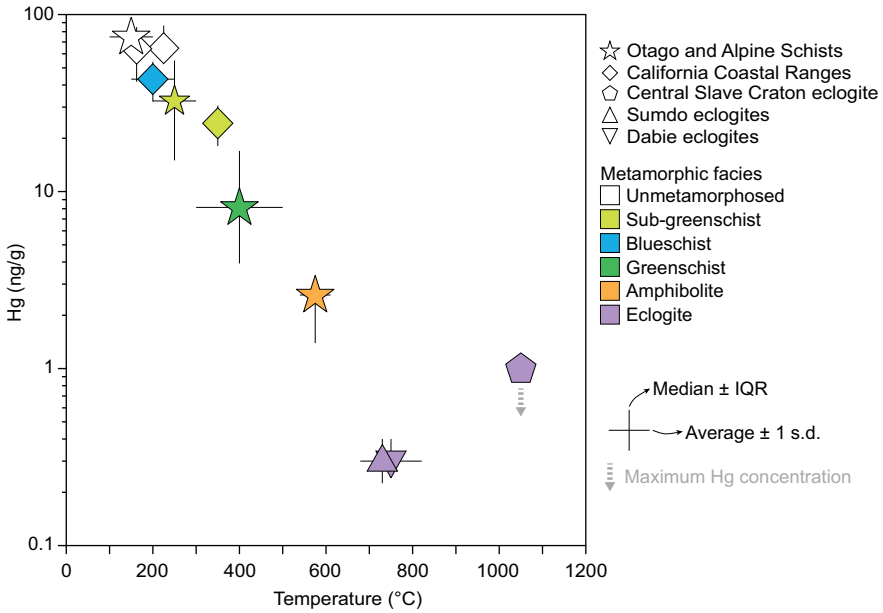

Figure 2 Mercury (Hg) concentrations (ng/g) *vs*. temperature (°C) in metamorphic samples from Otago and Alpine schists, New Zealand, California Coastal Ranges, USA, Dabie and Sumdo belts, China, and Central Slave Craton, Canada.

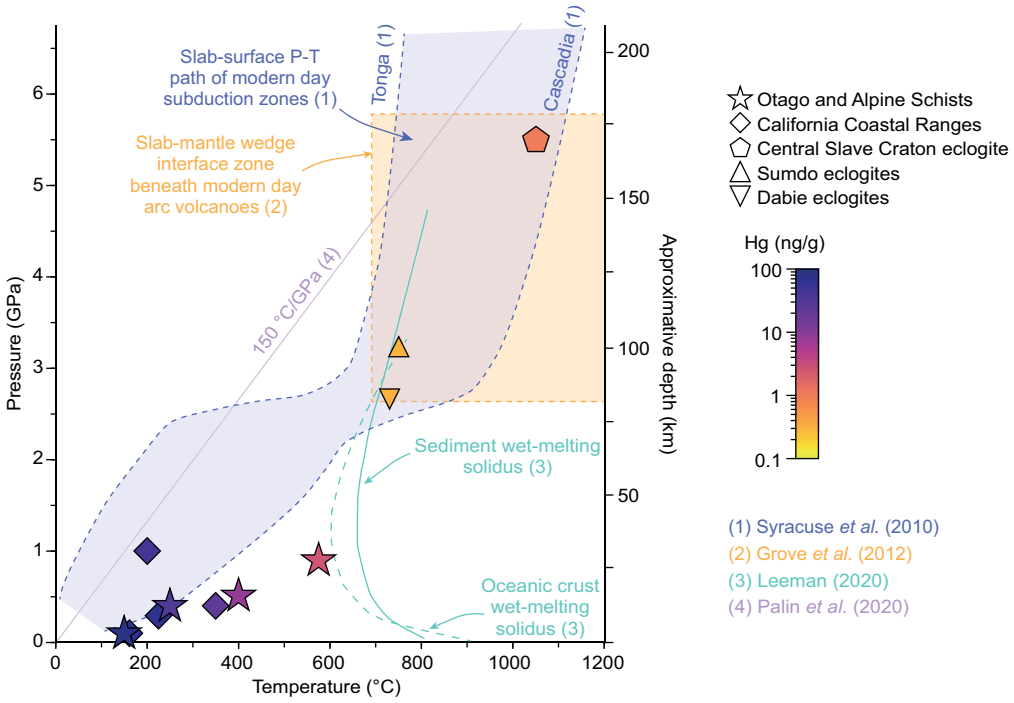

Figure 3 P-T diagram showing the devolatilisation of Hg. Mercury concentrations (ng/g) are median values discussed in the text. Tonga and Cascadia (Syracuse *et al.*, 2010) represent the coldest and hottest subduction zones (after Leeman, 2020). The geothermal gradient of 150 °C/GPa (Palin *et al.*, 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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Is Hg recycling into the Earth's mantle negligible?

F. Narduzzi, S. Covelli, M. Pistone, I. Pitcairn, R. Tribuzio, L. Ziberna

Supplementary Information

The Supplementary Information includes:

- Data Filtering
- Supplementary Figure S-1
- Metamorphic rocks
- Supplementary Figure S-2
- Supplementary Table S-1
- References, including those listed in supplementary Table S-1

Data Filtering

Literature data were filtered based on different geochemical parameters and observations. This allowed us to select the samples representing mantle-derived magmas with little or no evidence of differentiation and select the samples less affected by post-magmatic hydrothermal alteration. Analyses that either have been potentially affected by contamination or show low accuracy in the analysed standards, and samples for which no sufficient data were available to constrain the petrology and pressure-temperature conditions of equilibration, were also excluded.

Mantle-derived basalts and mafic rocks

Mercury analyses on igneous rocks linked to datasets where major elements were uncertain and/or not reported have been discarded. Also, all data on igneous systems associated with fractional crystallisation (FC) and/or assimilation

fractional crystallisation (AFC) (*e.g.,* Coufalick *et al.,* 2018) or interaction with crustal or seawater-derived fluids that boost Hg concentrations were removed from our analysis. To this goal, major elements of the remaining magmas were screened on the LOI-free basis, and only those samples showing Mg# (Mg/[Mg + Fe] mol%) ≥60 were considered. This cutoff helps decrease the possibility of an unrecognised small % of FC and/or AFC. Last but not least, data reporting Hg concentrations in cumulitic rocks were also excluded from our analysis.

Komati Formation komatiites. An interesting example of post-magmatic hydrothermal alteration is from the ~3.45-Ga-old komatiite pillow lavas of the Komati Formation (Barberton Greenstone Belt; Cloete, 1999). Instrumental Neutron Activation Analysis (INAA) was applied to determine Hg concentrations (118 ± 35 ng/g) in the cores of these high Mg# (>60) lavas. Contamination of the analyses due to the use of a jaw crusher before sample powdering appears less likely to explain such high values because the jaw crusher was pre-contaminated with the same komatiitic material (Cloete, 1999). These concentrations cannot reflect those of an Archean peridotitic mantle either, due to its elevated ambient temperature (~1500-1800 °C, *e.g.,* Palin *et al.,* 2020), which contrasts with the high volatility of Hg. Rather, such Hg might reflect interaction with Archean seawater or cryptic alteration. For this reason, komatiites were excluded.

Kimberlites, Kimberly, South Africa. Muramatsu (1983) analysed Hg concentrations in some kimberlites and in garnet-phlogopite harzburgite and eclogite xenoliths from South Africa. Kimberlites had $7.5^{+0.3}_{-0.8}$ ng/g of Hg, and garnet-phlogopite harzburgite and eclogite xenoliths had 5 and 5.5 ng/g of Hg, respectively. Analyses were performed using flameless atomic absorption spectroscopy (hereafter flameless AAS or CV-AAS), following Heinrichs (1975). The limit of detection of the CV-AAS is ~10 ng/g, as further supported by Garuti *et al.* (1984), who used flameless AAS to analyse Hg in peridotites and reported analyses below the detection limit for samples with Hg <10 ng/g. Consequently, the Hg concentrations ≤8 ng/g in Muramatsu (1983) are ambiguous. Heinrichs (1975) reports that if reagents such as nitric acid are purified before sample dissolution, the limit of detection can be as low as 0.1 ng/g. However, in Muramatsu (1983), there are no pieces of information on the reagents adopted, if they were purified before sample dissolution, if the Hg results were corrected for the Hg of the blanks, what standards were used, and if the Hg concentrations of the standards were consistent with those published in the literature. On the other hand, Heinrichs (1975) reported CV-AAS analyses for various reference materials, which are, however, most of the times not in agreement with the published RM Hg concentrations (see table 2 in Heinrichs 1975). Additionally, it appears that the analyses were not corrected for Hg in the blanks, as the Hg of the blanks was not reported. In summary, it is difficult to estimate the realistic uncertainties in Muramatsu's (1983) data and we refrain from using his dataset.

Peridotites from ophiolites, orogenic massifs and mantle xenoliths

The same geochemical filtering was also applied to some ultramafic samples from Balmuccia and Finero peridotite massifs (Ivrea-Verbano Zone, Alps, Italy), whose major element concentrations are unknown and/or their Hg enrichment (Narduzzi *et al.,* 2025) might be related to interaction with crustal fluids as seen for other chalcophile elements (*e.g.,* Sinigoi *et al.,* 2016; Wang *et al.,* 2018). Moreover, early Hg analyses by Garuti *et al.* (1984) on Balmuccia,

Baldissero, and Finero peridotites were discarded due to recurrent criticism regarding the accuracy of the results (see discussions in Palme and O'Neil, 2014; Canil *et al.,* 2015; Narduzzi *et al.,* 2025).

The Sahlabad meta-ophiolites (Hg = 30 - 70 ng/g; Aftabi Hossien and Zarrinkoub, 2013) and most of the harzburgites and peridotites from ophiolites of northern Canadian Cordillera (Hg = 0.9 - 4.7 ng/g; Canil *et al.,* 2015) were also discarded because Hg increases with loss of ignition (LOI up to 20 wt. %, our threshold was set to 2 wt. %), indicating low temperature overprint (*e.g.,* Canil *et al.,* 2015). This evidence is also supported by the negative correlation between LOI and S (21 - 146 µg/g) and positive trend between Cu/S ratios and Hg in Canadian ophiolite peridotites (Canil *et al.,* 2015).

Regarding the Canadian Cordillera orogenic massif peridotites, based on the high S contents (75 - 481 µg/g) in massif peridotites compared to the mantle xenoliths and ophiolitic peridotites (150^{+85}_{-13} and 63^{+37}_{-25} µg/g, respectively; c.f. Fig. 5 in Canil *et al.,* 2015), Canil *et al.* (2015) suggest that this would indicate refertilisation while still residing in the lithosphere, rather than high-temperature alteration. Therefore, Hg would also indicate metasomatism, which is consistent with the lack of correlation with high LOI (see Canil *et al.,* 2015).

Supplementary Figure

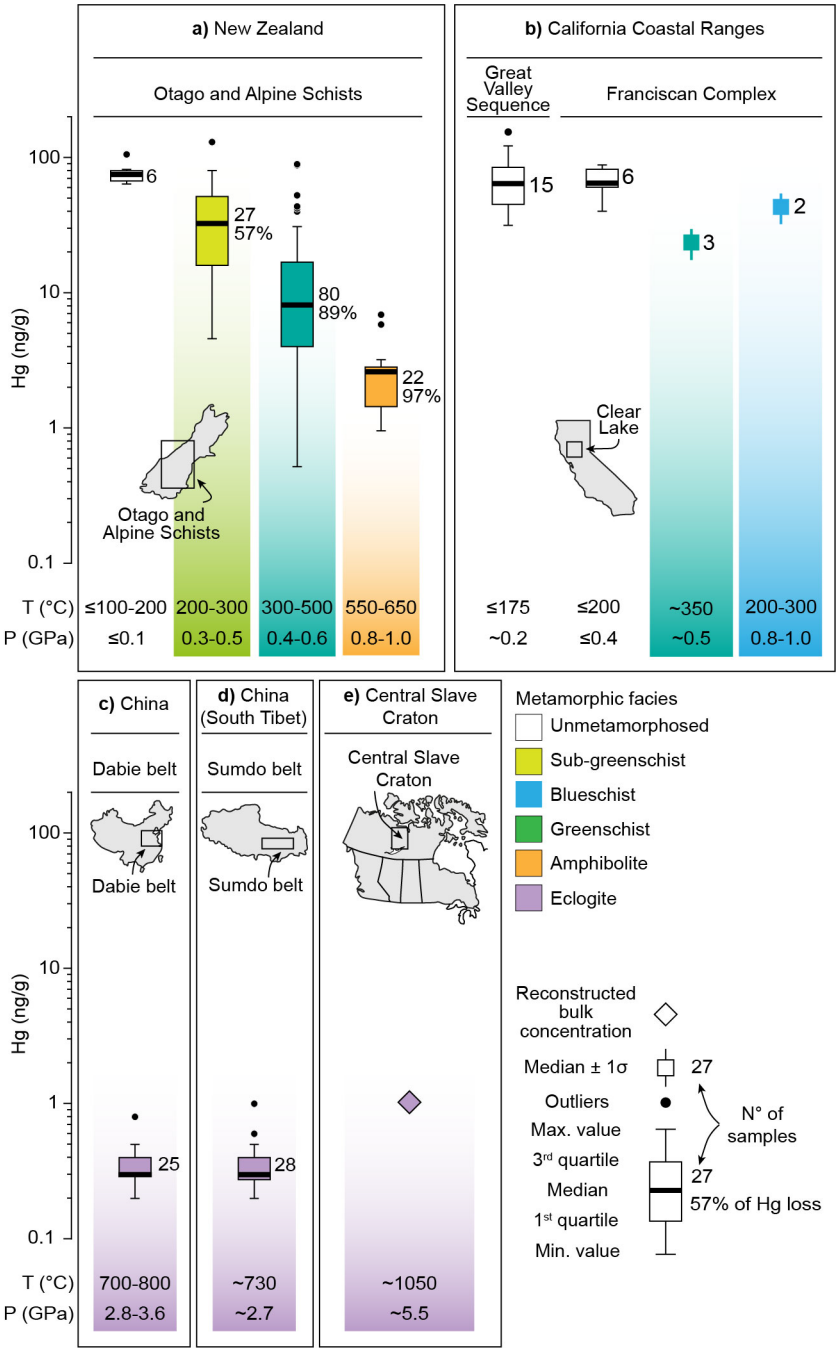

Figure S-1 Box plot diagram showing the Hg devolatilization in metamorphic terrains or belts. **a)** Otago and Alpine schist, New Zealand (Pitcairn *et al.,* 2006, 2010, 2014, 2015); **b)** California Coastal Ranges, USA (Smith *et al.,* 2008); **c)** Dabie and **d)** Sumdo belts, China (Chen *et al.,* 2025) and **e)** Central Slave Craton, Canada (Aulbach *et al.,* 2012). In b), the Franciscan complex also includes the serpentinites and metabasalts of the Coastal Ranges Ophiolite.

Metamorphic rocks

Here, we considered only those cases where Hg analyses are linked to quantitative P-T estimates, or the latter can be retrieved from other works. In the cases where this was not possible, Hg analyses were not considered further. For instance, Haas *et al.* (1984) measured Hg concentrations using CV-AAS technique but did not report the P-T conditions of the investigated metamorphic rocks. As the P-T conditions were impossible to retrieve from other works, these data were no longer considered. We also did not consider the Hg analyses of Hammerli *et al.* (2016) on the turbiditic metasediments of the Kanmantoo Group, Mount Lofty Ranges, South Australia. Mercury analyses were obtained on Li-borate fusion discs. That is, sample powders mixed with 12:22 borate flux at a sample to flux ratio of 1:6 were fused to glass after heating to 1050 ˚C. Such high temperatures could have caused Hg loss, thereby explaining the observed homogeneity across all samples despite different temperatures. The mercury analyses by Cameron and Jonasson (1972) are also doubtful because they may have been affected by unforeseen contamination.

Eventually, it is worth noting that Stepanov (2021) also discussed Hg data from neutron activation analyses of Marowsky and Wedephol (1971). Despite being seminal, Marowsky and Wedephol's (1971) work had significant uncertainties in the analysed standards. Figure S-2 reports all available Hg data published for the reference materials (RMs) used in the work of Marowsky and Wedepohl (1971). The majority of the data plotted in each diagram represents single Hg analyses of aliquots from the same bottle and from different bottles. On a few occasions, some data were reported as mean ± 1 standard deviation (1 SD; n analyses = 2-10). Each colour represents a different analytical technique. Marowsky and Wedepohl (1971) are shown separately to emphasise their data. For each of the RMs, we reported the median, the interquartile range (IQR; i.e., the 1st and 3rd quartiles), the average ± 1 SD and the RSD (%). The grey bar in each diagram represents the IQR. In these calculations, data from Marowsky and Wedepohl (1971) were not included to avoid potential bias. For each RM, the certified, proposed, and information values are also specified in red (Gonvindaraju 1994; Marie *et al*., 2015). For simplicity, we will refer to these values as reference values.

All RMs exhibit substantial Hg heterogeneity, likely due to powder heterogeneity within the same bottle and among different RM bottles (e.g., McNeal *et al.,* 1972; Cameron and Jonasson, 1972; Flanagan *et al.,* 1982; Marie *et al.,* 2015, among many). The high RSD calculated for each RM (35-63 %) supports RM heterogeneity. We also noticed that, except in the W-1 diabase diagram, NAA analyses from literature and Marowsky and Wedepohl (1971) always show lower and/or higher Hg concentrations compared to the Hg concentrations obtained by AAS and the other instruments. So, it is likely that RMs Hg variability is due to a combination of instrumental bias and sample inhomogeneity. Because the average and SD are sensitive to extreme values, we will use only the median and IQR.

Except for W-1 diabase, the reference values of the RMs (Gonvindaraju 1994; Marie *et al.,* 2015) are always below the median and mostly plot near the 1st quartile or, occasionally, outside the IQR (i.e., PCC-1 and G-1). Such a difference is observed even by excluding the NAA analyses. Although it is beyond the scope of this rebuttal letter to discuss the RMs reference values of Gonvindaraju (1994) and Marie et al. (2015), we suspect that these reference values are affected

either by undersampling or another type of bias. Note also that there are at least three plateaus at 170-180, ~110-130, and ~80-90 ng/g in W-1 Diabase and two plateaus at ~6-7 and ~33-35 ng/g in BRC-1 basalt. Other, less obvious plateaus can also be observed for the other RMs. Therefore, there might be at least two other potential "true values" for these two, excluding the possible true values we calculated here with the median. Therefore, obtaining a true value for these RMs is not trivial.

When considering the data from Marowsky and Wedepohl (1971), only the analyses for the W-1 and BCR-1 (Fig. 1) plot within the IQR and are close to the median and/or the RMs reference values. For PCC-1 RM, only one data point from Marowsky and Wedepohl (1971) is consistent with the reference value. Nonetheless, both are outside the IQR. The remaining data from Marowsky and Wedepohl (1971) lie outside the IQR and are therefore inconsistent with RM's reference values, the median, or both. Such inconsistency would persist even if the 10% analytical error reported by Marowsky and Wedepohl (1971) was taken into account. The offset from the median is 12 to 75 ng/g. The Hg analyses obtained for the W-1 diabase and BCR-1 basalt RMs (Fig. 1) are within the IQR, but given the observations reported above, this is a coincidence rather than a concordance. In summary, there are too many uncertainties and inconsistencies among the true Hg concentrations of the RMs, their reference values, our calculations, the NAA analytical uncertainties of Marowsky and Wedepohl (1971) and their data.

Regarding the data from the Swiss Alps, there are some points that must be considered, also considering the above observations. A literature search did not provide satisfactory results on the geology and petrography of the Keuper group (Frick quarry, northern Switzerland), from which the unmetamorphosed shales were collected. According to Marowsky and Wedepohl (1971), the shale sample with higher Hg (340 ng/g) appears to be 5 m above the other one with lower Hg (3 ng/g). That is, two unmetamorphosed samples from the same outcrop display an extreme variation of 335 ng/g in Hg over 5 meters. This is very suspicious. It is also suspicious that Hg concentrations in slightly metamorphosed (Hg = 3 ng/g) and greenschist rocks (Hg = 1-7 ng/g) are overall lower than those in amphibolite facies rocks (Hg = 9-14 ng/g). Although we cannot exclude a geological control, the pronounced divergence among NAA Hg concentrations from the literature and Marowsky and Wedepohl (1971), the other analytical methods, the RMs' reference values, and our medians is striking and cannot be ignored. As long as new Hg data on Swiss Alps samples are not provided, the Hg variability observed in Marowsky and Wedepohl (1971)'s dataset is most likely an analytical artefact.

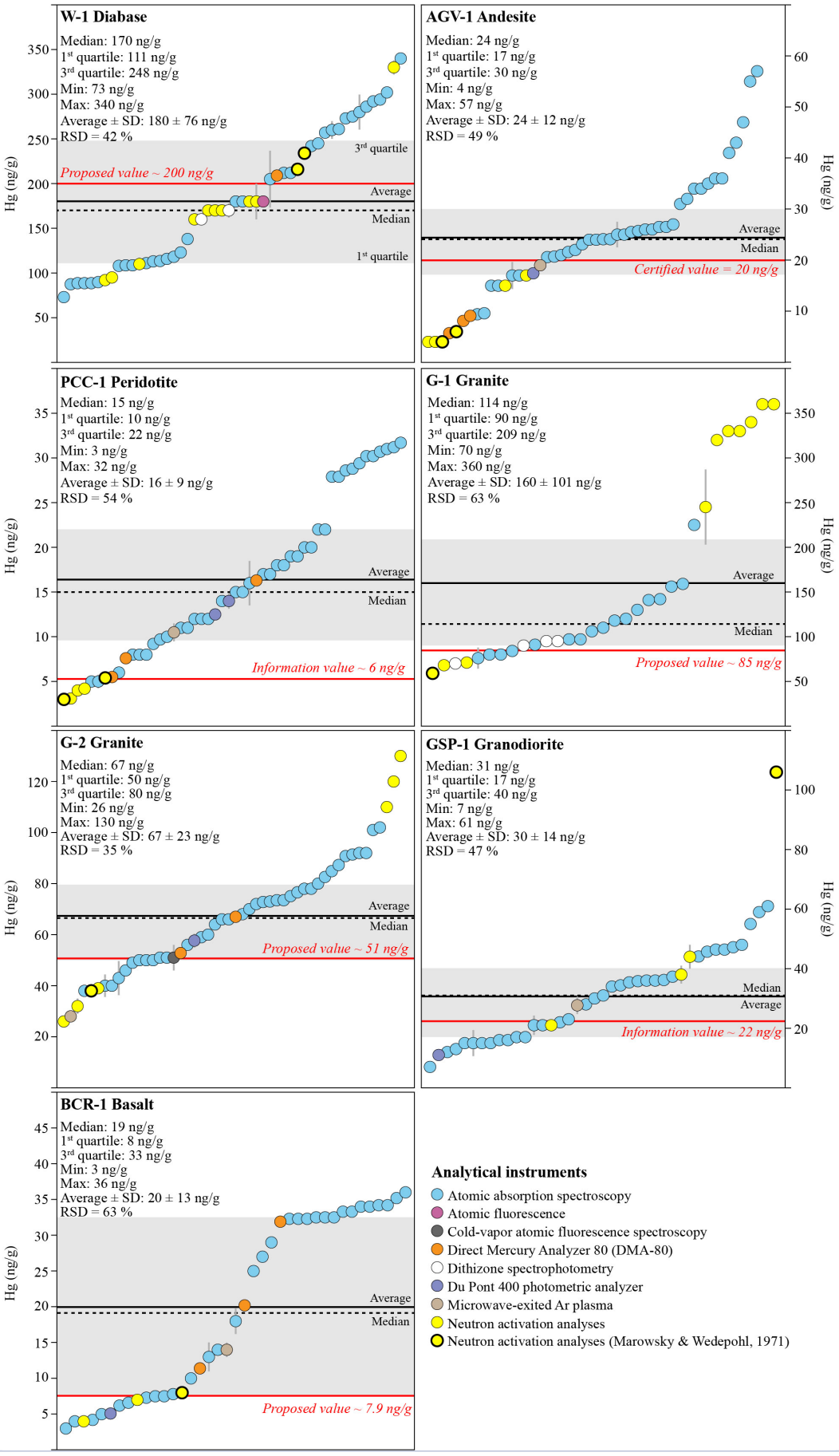
W-1 Diabase
Median: 170 ng/g
1st quartile: 111 ng/g
3rd quartile: 248 ng/g
Min: 73 ng/g
Max: 340 ng/g
Average ± SD: 180 ± 76 ng/g
RSD = 42 %
3rd quartile
Proposed value ~ 200 ng/g
Average
Median
1st quartile
AGV-1 Andesite
Median: 24 ng/g
1st quartile: 17 ng/g
3rd quartile: 30 ng/g
Min: 4 ng/g
Max: 57 ng/g
Average ± SD: 24 ± 12 ng/g
RSD = 49 %
Average
Median
Certified value = 20 ng/g
PCC-1 Peridotite
Median: 15 ng/g
1st quartile: 10 ng/g
3rd quartile: 22 ng/g
Min: 3 ng/g
Max: 32 ng/g
Average ± SD: 16 ± 9 ng/g
RSD = 54 %
Average
Median
Information value ~ 6 ng/g
G-1 Granite
Median: 114 ng/g
1st quartile: 90 ng/g
3rd quartile: 209 ng/g
Min: 70 ng/g
Max: 360 ng/g
Average ± SD: 160 ± 101 ng/g
RSD = 63 %
Average
Median
Proposed value ~ 85 ng/g
G-2 Granite
Median: 67 ng/g
1st quartile: 50 ng/g
3rd quartile: 80 ng/g
Min: 26 ng/g
Max: 130 ng/g
Average ± SD: 67 ± 23 ng/g
RSD = 35 %
Average
Median
Proposed value ~ 51 ng/g
GSP-1 Granodiorite
Median: 31 ng/g
1st quartile: 17 ng/g
3rd quartile: 40 ng/g
Min: 7 ng/g
Max: 61 ng/g
Average ± SD: 30 ± 14 ng/g
RSD = 47 %
Median
Average
Information value ~ 22 ng/g
BCR-1 Basalt
Median: 19 ng/g
1st quartile: 8 ng/g
3rd quartile: 33 ng/g
Min: 3 ng/g
Max: 36 ng/g
Average ± SD: 20 ± 13 ng/g
RSD = 63 %
Average
Median
Proposed value ~ 7.9 ng/g
Hg (ng/g)
Analytical instruments
Atomic absorption spectroscopy
Atomic fluorescence
Cold-vapor atomic fluorescence spectroscopy
Direct Mercury Analyzer 80 (DMA-80)
Dithizone spectrophotometry
Du Pont 400 photometric analyzer
Microwave-exited Ar plasma
Neutron activation analyses
Neutron activation analyses (Marowsky & Wedepohl, 1971)

Figure S-2 Compilation of Hg concentrations for the reference materials analysed in Marowsky and Wedepohl (1971). Data source: Morris and Killick (1964), Fleischer (1965, 1969, 1970), Alian and Shabana (1967), Ehmann and Lovering (1967), Hatch and Ott (1967), Ishida *et al.* (1970), Laul *et al*. (1970), Marowsky and Wedepohl (1971), Muscat and Vickers (1971), Omang and Paus (1971), Weissberg (1971), Cameron and Jonasson (1972), McNeal *et al*. (1972), Corte and Dubois (1975), Heinrichs (1975), Azzaria and Carrier (1976), Casas and Vaquer (1979), Srivastava (1979), Sighinolfi *et al*. (1980), Flanagan (1982), Kennedy and Crock (1987), Hageman (2007), Marie *et al*. (2015). Certified, proposed, and information values of the reference materials are from Govindaraju (1994) and Marie *et al*. (2015).

California Coastal Ranges

Smith *et al.* (2008) did not report the P-T peak conditions of the metamorphic rocks they analysed. However, as their rocks belong to the Goat Mountains (Clear Lake district, California), P and T can be retrieved from other works discussing the metamorphism of this area:

- Great Valley (Clear Lake district, California) sediments are weakly metamorphosed to the zeolite facies at a temperature of ≤250 °C (Blake *et al.,* 1988, cited in Smith, 2010). The same sediments were analysed for their oxygen isotopes, and it was found that T did not exceed 175 °C, which is consistent with burial metamorphism (Sucheki and Land, 1983). Sucheki and Land (1983) also report that the sediments were buried at depths of 6 – 7 km, corresponding to a geotherm of ~25 °C/km, from which it can be calculated a T between 150 and 175 °C. According to these data, we calculated a P of ~0.1 GPa for the Great Valley sediments;

- Franciscan Complex sediments have been metamorphosed to a maximum of pumpellite-prehnite facies with a T of ~250 °C (Ernst *et al.,* 1970 in Smith 2010). Wakabayashi *et al.* (2010) suggest a T <200 °C and P ≤0.4 GPa for rocks of the Goat Mountains undergoing zeolite to pumpellite-prehnite facies. From these data, we use a T of ~ 225 ± 35 °C and a P of ~0.3 GPa, which also represents the transition between the two metamorphic facies;

- Franciscan blueschists of the Goat Mountains were investigated by Ernst *et al.* (1970), who proposed a recrystallisation T of 200 to ≥ 300 °C (350 °C) and P of up to or exceeding 0.8 GPa. Blueschists in this area have been mapped by Wakabayashi (2017) as non-schistose blueschist facies, with some inclusions of lawsonite–albite facies. Wakabayashi *et al.* (2010) reported that lawsonite blueschists exhibit a T of 150 – 250 °C and P from ~0.6 to ≥1 GPa. If a T average of 250 ± 50 °C is taken and considering a maximum P of ~1.3 GPa which indicates the beginning of the forbidden zone at this temperature (see Palin *et al.,* 2020 for example), and the minimum P of 0.6 GPa, a P average of 1.0 ± 0.5 GPa is obtained. According to these considerations we T of 200 ± 50 °C and P of 1.0 ± 0.5 GPa;

- Data on the P-T conditions of the Coastal Ranges Ophiolites metabasalts and serpentinites (Smith *et al.,* 2008; Smith, 2010) are rather scanty. Wakabayashi (2017) reports that these rocks underwent sea-floor hydrothermal metamorphism ranging from zeolite to greenschist or higher grade, but burial metamorphism does not exceed zeolite grade. Since no other information is reported, we use an average T of ~350 °C and P of ~0.4 GPa corresponding to P-T average values for greenschist metamorphic facies.

Supplementary Table

Table S-1 (.xlsx) is available for downloading from the online version of this article at https://doi.org/10.7185/geochemlet.2626

Table S-1 Mercury (Hg) and major element concentrations of the samples used in this study. Details of the pressure and temperature conditions of the metamorphic rocks are also reported. Further details can be found directly in the spreadsheet.

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