






## RESEARCH ARTICLE

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# Global Hydrogen Production During High-Pressure Serpentinization of Subducting Slabs

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### Key Points:

- First-order estimate of the gross flux of “slab-serpentinization” and the resulting (possible) hydrogen production (4.2–24 • 10<sup>7</sup> kg of H<sub>2</sub> per annum)
- Seafloor spreading history and ocean basin evolution (prior to subduction) impart the strongest control on slab-serpentinization possibility

### Supporting Information:

Supporting Information may be found in the online version of this article.

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**Abstract** Serpentinization is among the most important, and ubiquitous, geological processes in crustal–upper mantle conditions (<6 GPa, <600°C), altering the rheology of rocks and producing H<sub>2</sub> that can sustain life. While observations are available to quantify serpentinization in terrestrial and mid-ocean ridge environments, measurements within subduction zone environments are far more sparse. To overcome this difficulty, we design a methodology to quantify and offer a first-order estimate of the magnitude of “slab-serpentinization” that has occurred over the last 5 Ma within the world’s subduction zones by coupling four discrete tectonic and geophysical datasets—(a) raster grids of relic abyssal peridotite (peridotite exhumed from slow spreading mid-ocean ridges but unaffected by pre-subduction serpentinization) within ocean basins, (b) slab geometry, (c) thermal profiles and a (d) plate-tectonic model. Averaged per year, our results suggest that 4.2–24 • 10<sup>7</sup> kg of H<sub>2</sub> per annum could be generated from “slab-serpentinization” within a subduction zone. Our estimate is 3–4 orders of magnitude lower than what is thought to be produced at mid-ocean ridges, and 1–2 orders of magnitude lower than what could occur through serpentinization at trench flexure and when including possible mantle wedge serpentinization. Higher hydrogen production is correlated most strongly with the spreading history of ocean basins, underlaying the importance of the tectonic history of a slab prior to subduction.

**Plain Language Summary** The fate of most ocean crust formed at mid-ocean ridges is to eventually subduct and be recycled into the mantle. Subduction zones therefore represent a key link between the rocks we see at the surface of the Earth, both in oceans and continents, and the underlying mantle. However, subduction zones are impossible to observe directly and therefore difficult to fully understand the processes that shape them. Here, we designed a framework that coupled a series of discrete data sets to model how the composition of each subducting slab across the globe differs in order to provide an accurate estimate of “slab-serpentinization.” Serpentinization is the process that converts mantle rocks to serpentinite through exposure to water. A by-product of this process is the formation of hydrogen gas. Using our framework, we estimated bulk fluxes of serpentinization in subducting slabs, and the corresponding flux of hydrogen.

## 1. Introduction

The evolution of aqueous fluids during subduction zone metamorphism plays a central role in global cycling of life-essential elements, their redox state, and their return to the Earth’s surface (Manning and Shock, 2013; Schmidt & Poli, 2013; Sverjensky & Daniel, 2020). Based on the chemistry of arc magmas, the redox state of forearc-to-subarc slab (50–100 km depth) fluids has long been considered oxidized and dominated by aqueous fluids released through the dehydration of the subducting slab rich in CO<sub>2</sub> (e.g., Debret & Sverjensky, 2017; Padrón-Navarta et al., 2023; Parkinson & Arculus, 1999; Tumiami & Malaspina, 2019). However, several studies over the last 15 years have highlighted the possibility for more reduced fluids, rich in H<sub>2</sub> and other reduced species, and reduced assemblages to form in subduction zone conditions ranging from forearc to sub-arc depth (e.g., Arai et al., 2012; Boutier et al., 2021; Debret et al., 2022; Ferrando et al., 2010; Galvez et al., 2013; Huang et al., 2017; Kutcherov et al., 2020; Malvoisin et al., 2011; Piccoli et al., 2019; Sachan et al., 2007; Tao et al., 2018;

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Vitale Brovarone et al., 2017, 2020). Assessing the magnitude of deep H<sub>2</sub> in subduction zones is crucial because the presence of such reduced fluids may strongly affect current models of deep geochemical cycling and could act as a source of energy for the deepest subsurface microbial life (Plümper et al., 2017; Rogers et al., 2023; Vitale Brovarone et al., 2020).

The aqueous alteration of ultramafic rocks, or serpentinization, is a key process in forming H<sub>2</sub> and promoting reducing conditions (Andreani et al., 2013; Janecky & Seyfried, 1986). Serpentinization is readily observed in natural environments: at mid-ocean ridges (e.g., Charlou et al., 2002; Kelley et al., 2005; Liu et al., 2023) and on land (e.g., Barnes et al., 1978; Leong et al., 2021; Leong & Shock, 2020; Miller et al., 2016; Sánchez-Murillo et al., 2014), requiring only peridotite to be exposed to water at crustal-to-upper-mantle conditions (<~6 GPa, <~600°C) (Janecky & Seyfried, 1986). The process has also been recreated within laboratory experiments with both natural and synthetic samples at a range of pressures and temperatures (see recent review by Barbier et al., 2020) and thermodynamic modeling of subduction zones predicts the presence of serpentinite minerals, chiefly antigorite, up to a depth of 6 GPa (Abers et al., 2017; Maurice et al., 2020; Ulmer & Trommsdorff, 1995) (though H<sub>2</sub> could be generated more deeply as olivine reacts with water to form magnetite and H<sub>2</sub>, e.g., Malaspina et al., 2023).

Geological evidence of serpentinization within subduction zones is more sparse, despite geophysical evidence suggesting it could affect substantial volumes of convergent margins (Blakely et al., 2005; Bostock et al., 2002; Ranero et al., 2003; Williams & Gubbins, 2019). Above subducting slabs, serpentinization may affect the so-called “cold-nose” of the fore-arc mantle wedge (Abers et al., 2017; Blakely et al., 2005; Bostock et al., 2002; Debret et al., 2022; Fryer, 1996; Hyndman & Peacock, 2003; Mottl et al., 2003; Ohara et al., 2012; Plümper et al., 2017; Vitale Brovarone et al., 2020). Within subducting slabs, serpentinization may take place by trench flexure (e.g., Faccenda, 2014; Ranero et al., 2003), or deeper in subduction zones through the interaction between metamorphic fluids and relic (or residual) abyssal peridotite (RAP)—i.e. peridotite that was exhumed, but not fully serpentinized, at slow spreading ridges and is transported via seafloor spreading to subduction zones and undergoes serpentinization during subduction (Figure 1c, Boutier et al., 2021; Vitale Brovarone et al., 2020).

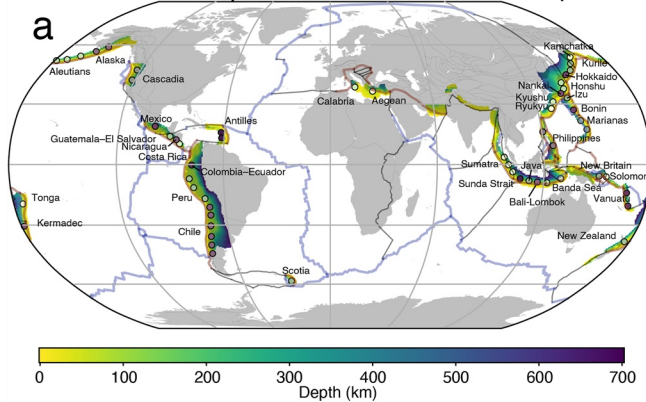
This process of “slab-serpentinization” in the upper most 7 km of the subducting slab (corresponding to the alteration depth of thinned crust at mid-ocean ridges, e.g., Cannat et al., 2010; Merdith et al., 2020) is directly constrained by the raw volume of reactant (i.e., RAP) available, and the pressure-temperature conditions of a subduction zone that control the breakdown of hydrous minerals. For this study, we are interested in the “slab-serpentinization,” which is defined by the pressure-temperature stability curve of antigorite, the highest-temperature serpentinite mineral stable at high-pressure conditions in subduction zones (e.g., Evans, 2004; Schwartz et al., 2013). While some estimates of serpentinization exist from hydration of the forearc mantle wedge (e.g., Carlson & Miller, 2003; Hyndman & Peacock, 2003; Williams & Gubbins, 2019), the presence and extent of slab-serpentinization within subduction zones remain much more speculative (e.g., Vitale Brovarone et al., 2017). Consequently, there is currently no volumetric estimate of possible slab-serpentinization within subduction zones or of the resulting hydrogen flux.

In this study, we approach the problem by coupling discrete data sets including slab geometry (Hayes et al., 2018), thermal profiles (Syracuse et al., 2010) and subduction histories of the world's subduction zones (Müller et al., 2016) (Figures 1a and 1b; Figures S1–S3 in Supporting Information S1). We do this to generate estimates of the global amount of RAP entering subduction zones as part of the subducting oceanic lithosphere (Merdith et al., 2019, 2020). Assuming the sufficient availability of aqueous fluids in the subducting plate (e.g., Faccenda, 2014; van Keken et al., 2011), we estimate the global volume of slab serpentinization and resulting hydrogen production, arising from the upper 7 km of the subducting slab. Our analysis does not consider serpentinization of the mantle wedge and deliberately excludes possible serpentinization of the lithospheric mantle due to trench flexure, the later process which may lower the amount of available RAP for slab-serpentinization, but promote additional serpentinization and H<sub>2</sub> production at convergent margins.

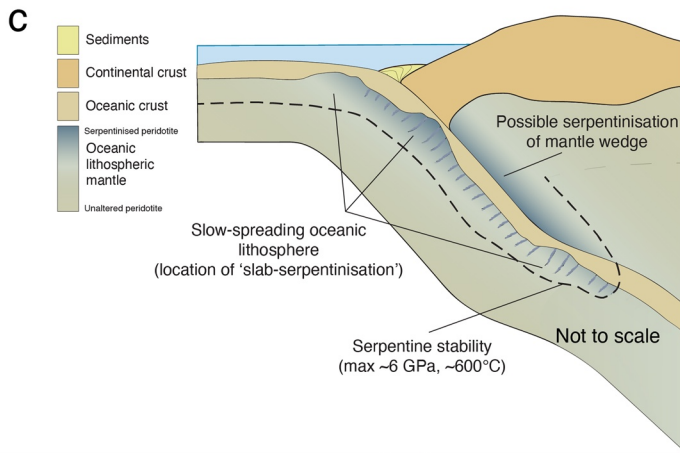
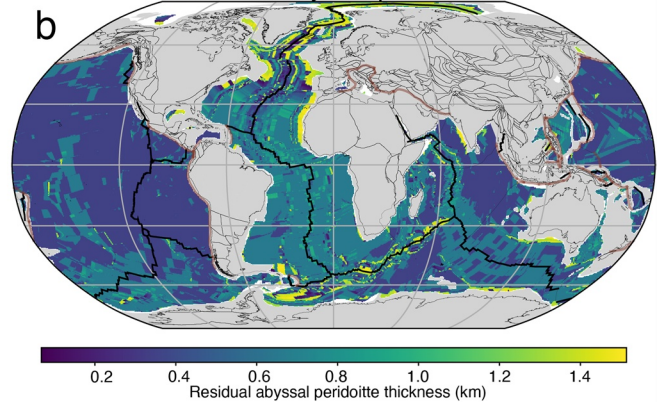
## 2. Overview of Our Approach

Recently, a number of studies have developed techniques to map features forward in time, from ocean basins onto subducted slabs (Harmon et al., 2019; Hicks et al., 2022; McGirr et al., 2021). Here we extend the approach of McGirr et al. (2021) by forward propagating time-sensitive rasterized-grids of different ocean crust properties

Subducted slab depth and locations of thermal profiles



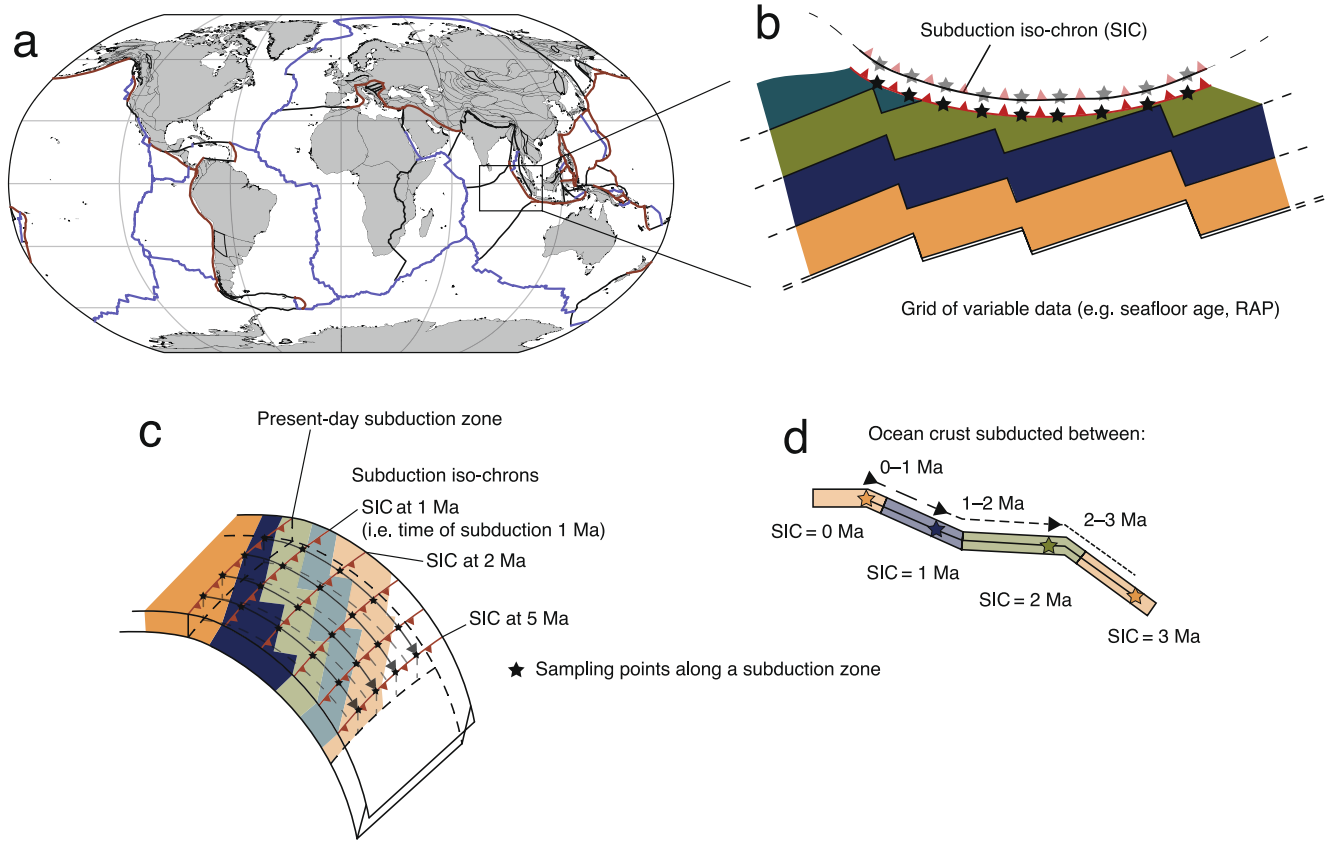
Residual abyssal peridotite



**Figure 1.** (a) Map of Slab2 and Syracuse et al. (2010) data sets. Each circle represents a sample site of thermal properties from Syracuse et al. (2010). (b) Residual abyssal peridotite grid at 0 Ma after Merdith et al. (2019, 2020); (c) Schematic diagram of a subduction zone highlighting “slab-serpentinization” relative to other established settings of serpentinization at subduction zones.

(e.g., seafloor age, RAP content) into a subduction zone (Figure 2, see Supporting Information for more details). We do this forward propagation using the kinematic history of the ocean basins modeled with full-plate reconstructions (models that explicitly model the kinematic evolution of tectonic plates and plate boundaries, e.g., Seton et al. (2023)) to estimate the forward evolution of an ocean plate once it intersects a modeled subduction zone and begins subducting.

We begin our analysis 5 Ma ago to limit our study to the times when the subduction history of the plate model (Müller et al., 2016) is best constrained, though in principle the method could be applied to earlier times. At the start time (i.e., 5 Ma), we extract the velocity orientation and convergence rate of a subducting plate at equally spaced points (latitude, longitude) along the subduction zone. Each point is also used to extract relevant tectonic parameters from our seafloor grids (where they intersect with the subduction zone), including RAP content, seafloor spreading rate at the formation and age of oceanic crust (in principle, any feature intersecting a subduction zone can be used in our method). At each point, we then calculate a dip angle following Mather et al. (2023), who approximate dip based on a range of subducting and over-riding plate properties including age, thickness, buoyancy and rollback (Hu & Gurnis, 2020). The equally spaced points along the subduction zone, therefore, represent the initiation of a *hypothetical subduction isochron*—a line drawn along a subducting slab representing the same time of subduction (e.g., Figure 2, analogous to a seafloor spreading isochron). We can then calculate, using these parameters, the position of the points (latitude, longitude, and depth, using dip angle) and intersected tectonic features after 1 Ma (i.e., 4 Ma ago). We repeat this process for new points intersecting subduction zones, using the new trench-values of orientation, convergence rate and dip to propagate both the new



**Figure 2.** Schematic summary of our approach. (a) Plate tectonic model at 0 Ma, highlighting plate boundaries. (b) Sampling at regular intervals across a subduction zone, with artificial isochrons highlighted (the black stars represent the points we sample the hypothetical ocean-basin raster before subduction). (c) Block diagram of a subduction zone showing “subduction isochrons” which the points we subduct represent. (d) As in (c), but a cross section view.

points as well as the previously “subducted” points, forward. This process is repeated until we reach the end time (in this analysis at 0 Ma).

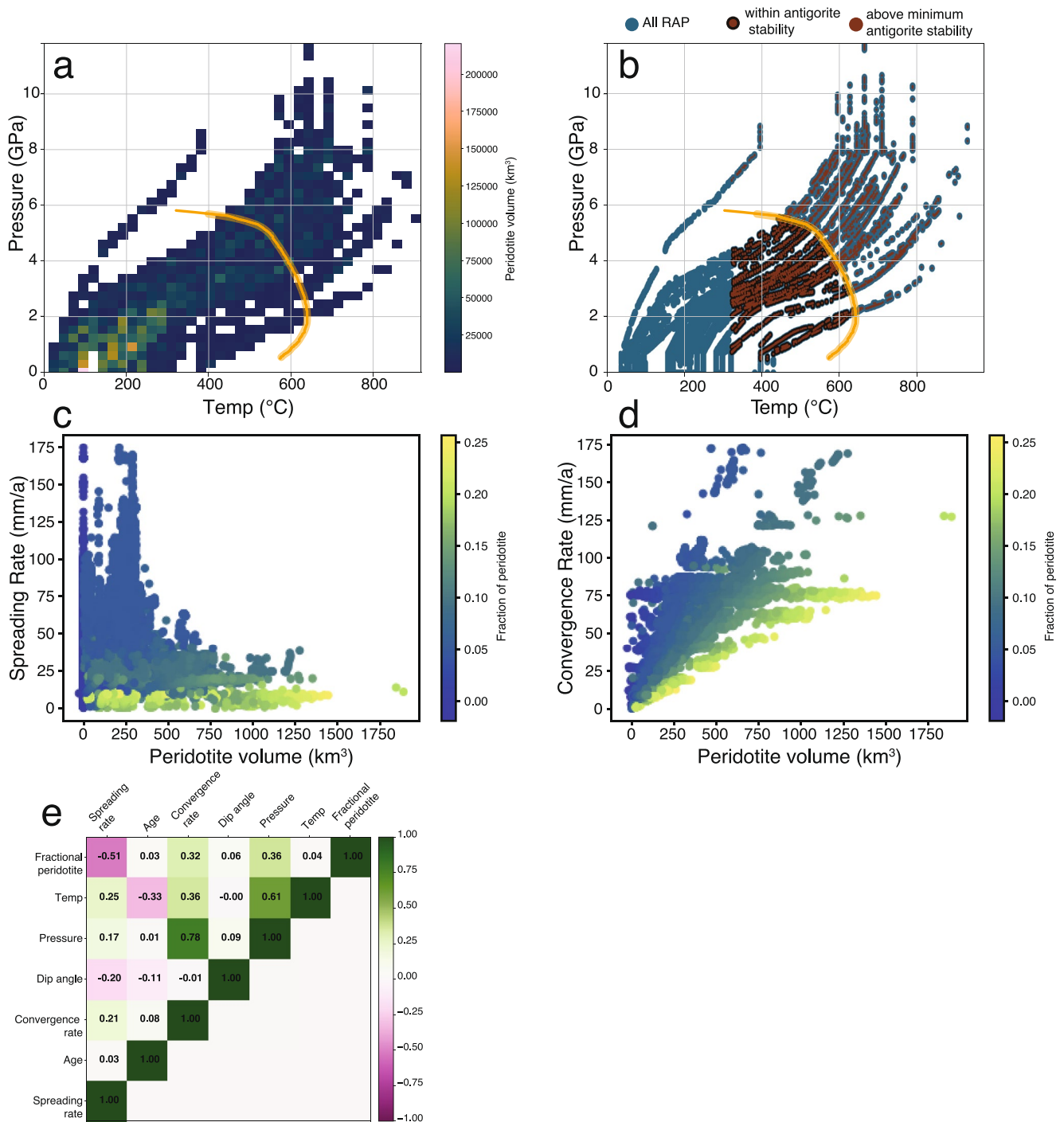
To generate some idea of uncertainty on our method and calculation, we also performed the analysis without correcting for dip. For this second analysis, we also calculated distance from the present-day trench for each point at 0 Ma once the analysis had finished (i.e., based on the convergence rate how far has each point moved since it was “subducted”). We then used this distance to calculate the corresponding depth in the Slab2 geometry based on the distance along the slab surface from the present-day trench. This approach assumes that in the shallow parts of the subduction zones over 1 Ma timescales, slab motion is accounted for by the motion of the downgoing plate.

In order to match our points with the thermal profiles of Syracuse et al. (2010) (SYR10, Figure 1a), we find the nearest geographic profile to each of our subducted points. Because each point that we have subducted contains a “depth” estimate (either through our dip approximating, or directly associating distance from trench to depth along a Slab2 profile), we extract the temperature from the corresponding SYR10 profile at the corresponding depth of our subducted point at the slab Moho (i.e., the temperature between the slab-crust and slab-mantle). Finally, we calculated lithostatic pressure at the modeled point depth.

### 3. Results

Figure 3 displays the sum of all RAP subducted in the last 5 Ma, with the antigorite stability field depicted at varying pressure-temperature conditions (orange line, Figure 3a). Our results are presented in three formats, first with respect to the total amount of RAP subducted over the last 5 Ma (blue circles in Figure 3b), second, with the RAP that sits within the antigorite stability field at present-day (red circles with black outlines in Figure 3b),





**Figure 3.** Summary of results, (a, b) distribution of RAP in global subduction zones within the antigorite stability field (orange line approximates the lower stability band of antigorite (Schwartz et al., 2013)). Scatterplots highlighting the relationship between the volume of peridotite in subduction zones with spreading rate (c) and convergence rate (d). (e) Pearson correlation coefficient matrix for tectonic parameters—fractional peridotite refers to the fraction of the upper 7 km of the slab that is RAP and the conversion of peridotite mass to hydrogen production is linear. Results use a representative H<sub>2</sub> production value of 200 g per m<sup>3</sup> of reactant.

and finally, with respect to the amount of RAP that sits within, or has passed through, the antigorite stability field at present-day (i.e., including RAP that has been subducted past the upper stability of antigorite, all red circles (including those with black outlines) in Figure 3b).

We calculate a total RAP volume subducted in the last 5 Ma of  $9.2 \cdot 10^6$  km<sup>3</sup> (Figure 3a), with approximately 7%–20% of this volume ( $7$ – $18 \cdot 10^5$  km<sup>3</sup>) currently located within the antigorite stability field (Figure 3a, orange

**Table 1**

Summary of Results of Total Fluxes of RAP Being Subducted and Possible Hydrogen Generation Through Serpentinization

RAP	All subducted RAP (all points in Figure 3b)			Currently within antigorite stability (red points with black outlines in Figure 3b)			Above minimum antigorite stability (0.5 GPa, 320°C, red points in Figure 3b)		
	Moles	Volume (km <sup>3</sup> )	Mass (kg)	Moles	Volume (km <sup>3</sup> )	Mass (kg)	Moles	Volume (km <sup>3</sup> )	Mass (kg)
Per 5 Ma	9.2 • 10 <sup>6</sup>	3.0 • 10 <sup>19</sup>	3.0 • 10 <sup>19</sup>	7–18 • 10 <sup>5</sup>	2.3–6.0 • 10 <sup>18</sup>	2.3–6.0 • 10 <sup>18</sup>	2.3–4.1 • 10 <sup>6</sup>	7.7–13 • 10 <sup>18</sup>	7.7–13 • 10 <sup>18</sup>
Average per annum	1.8	6.1 • 10 <sup>12</sup>	6.1 • 10 <sup>12</sup>	0.13–0.36	5–12 • 10 <sup>11</sup>	5–12 • 10 <sup>11</sup>	0.5–0.8	1.5–2.7 • 10 <sup>12</sup>	1.5–2.7 • 10 <sup>12</sup>
H <sub>2</sub> Per 5 Ma	4.1–14 • 10 <sup>14</sup>	8.3–28 • 10 <sup>14</sup>	8.3–28 • 10 <sup>14</sup>	3.1–27 • 10 <sup>13</sup>	6.2–54 • 10 <sup>13</sup>	6.2–54 • 10 <sup>13</sup>	1.0–6.0 • 10 <sup>14</sup>	2.1–12 • 10 <sup>14</sup>	2.1–12 • 10 <sup>14</sup>
Average per annum	8.2–27 • 10 <sup>7</sup>	1.7–5.5 • 10 <sup>8</sup>	1.7–5.5 • 10 <sup>8</sup>	6.1–54 • 10 <sup>6</sup>	1.2–11 • 10 <sup>7</sup>	1.2–11 • 10 <sup>7</sup>	2.1–12 • 10 <sup>7</sup>	4.2–24 • 10 <sup>7</sup>	4.2–24 • 10 <sup>7</sup>
Fraction of total			0.07–0.2						0.25–0.44

bounds, Figure 3b). However, this estimate omits RAP that has already passed through the antigorite stability field. Including these (now) more deeply subducted sources (red points, Figure 3b), the gross flux of RAP that underwent serpentinization to antigorite is 2.3–4.1 • km<sup>3</sup> over the last 5 Ma; this represents 25%–44% of all subducted RAP and gives an averaged flux of 0.5–0.8 km<sup>3</sup> per annum (Table 1).

To calculate hydrogen production, we follow the approach of Vitale Brovarone et al. (2020), who suggested that 90–300 g of H<sub>2</sub> is produced per 1 m<sup>3</sup> of reactant peridotite assuming ~50% serpentinization and a fluid-rock ratio of 1:1 (figures are produced using a representative value of 200 g per 1 m<sup>3</sup>, see SI for further details). This method yields a total hydrogen production (from peridotite within and that has passed through the antigorite stability field at present-day) of 2.1–12 • 10<sup>14</sup> kg (1.0–6.0 • 10<sup>14</sup> mol) of H<sub>2</sub>, roughly 4.2–24 • 10<sup>7</sup> kg (2.1–12 • 10<sup>7</sup> mol) of H<sub>2</sub> per annum. A “present-day” flux based on our modeled RAP currently within the antigorite stability field is the same order of magnitude, between 1.2 and 11 • 10<sup>7</sup> kg (6.1–54 • 10<sup>6</sup> mol) of H<sub>2</sub>.

To investigate H<sub>2</sub> production within different subduction zones, we isolated the volume of subducted peridotite against both convergence rate (at time of subduction) and spreading rate (at time of seafloor production) (Figures 3c and 3d). As well as calculating a Pearson's Correlation matrix to understand the relationship between various tectonic parameters and the amount of hydrogen formed (which is a linear product of peridotite content) (Figure 3e). Our results indicate that the strongest positive correlation is between peridotite, and pressure or depth and convergence rate (both 0.36 and 0.32 respectively), with the relationship between peridotite volume and convergence rate also clear in Figure 3d. These positive correlations are likely because a high convergence rate results in a larger overall volume of subducted oceanic lithosphere at greater depths. There is a strong negative correlation between spreading rate at the time of formation and fractional peridotite content (–0.51), as expected (also Figure 3c). The correlation between spreading rate and RAP becomes more strongly negative (–0.73) if we consider just the thickness of peridotite being subducted, and more weakly negative (–0.03) if we consider its absolute volume. The reason for the weakening of the correlation when considering the volume of RAP is that currently the majority of subduction occurs around the intermediate–fast spreading Pacific Ocean, thus the absolute volume of crust being subducted outweighs the lower amount of modeled RAP preserved in the crust.

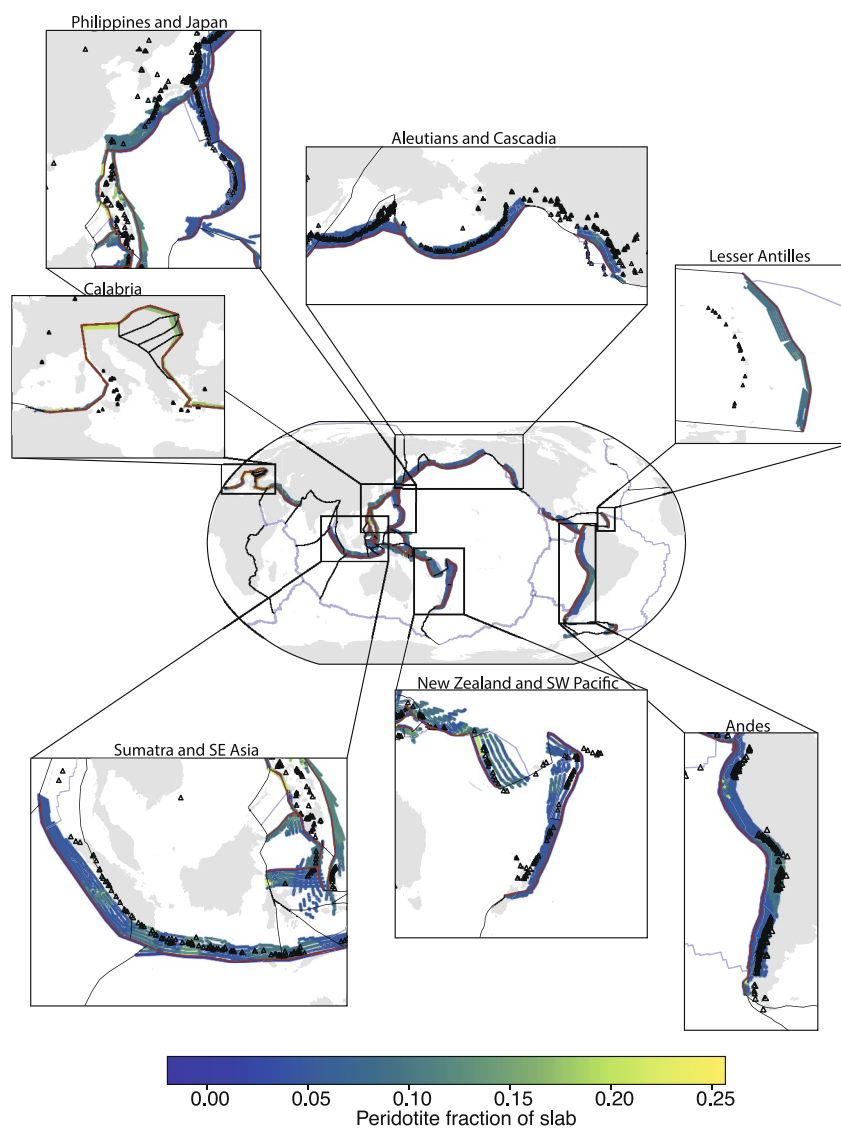
## 4. Discussion

### 4.1. Tectonic Controls on Slab-Serpentinization

Our analysis does not include any estimate of the serpentinization taking place within the seafloor, after mid-ocean ridge serpentinization and before it enters the subduction trench. Therefore, the kinematic conditions of the ridge segment where our subducting lithosphere originally formed exert a first-order control on the amount of available RAP. This is because ocean lithosphere formed at ultra-slow–slow-spreading ridges contains significant volumes of exhumed mantle (e.g., Tucholke & Lin, 1994), and when this lithosphere eventually subducts, there are large volumes of relic peridotite (relative to crust at a fast-spreading ridge) available to be serpentinized within the top 7 km. Broadly, the method we have implemented (Merdith et al., 2020) suggests that on average about 1/3rd of new oceanic lithosphere at a slow–ultraslow spreading ridge are mafic volcanics and 2/3rds are exhumed peridotite, with approximately half of the exhumed peridotite reacting to form serpentinite. This estimate is in line with other estimates of exhumation and serpentinization of mid-ocean ridge peridotites (Cannat et al., 2010; Worman et al., 2016).

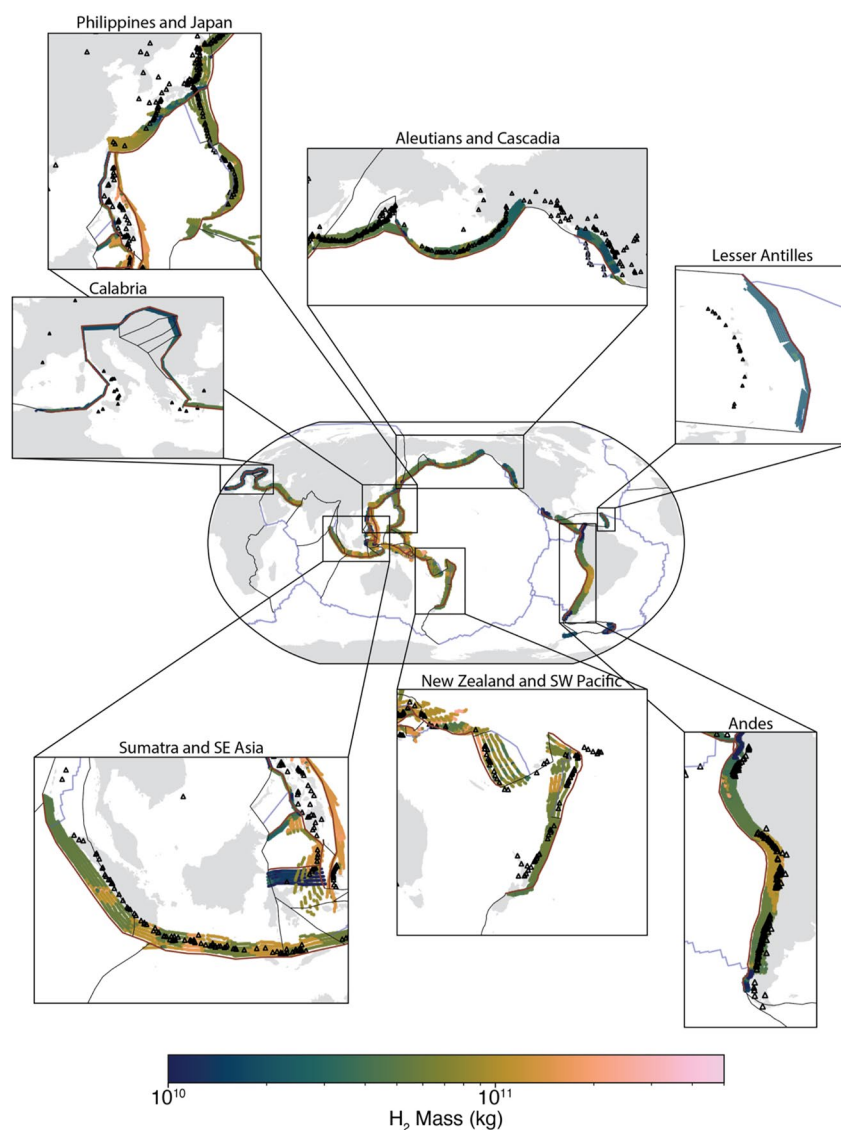
The influence of the seafloor spreading history can be seen in a map-view of our results (Figures 4 and 5). For example, the Andean and Izu-Bonin systems produce significantly less H<sub>2</sub> than other subduction zones such as the Philippines or Sumatra. This is a direct consequence of the fact that the seafloor being subducted at the Andean margin formed at an intermediate–fast spreading ridge (Pacific-Farallon ridge, e.g., Seton et al., 2020, 2012) and is unlikely to contain large amounts of exhumed abyssal peridotite. Conversely, in the north-eastern Indian ocean, segments of the ocean crust being subducted under Sumatra formed at a modeled ultraslow–slow ridge (e.g., Jacob et al., 2014), and the volume of peridotite and resulting hydrogen are inferred to be three–four times larger (Figures 4 and 5).

Complementing the seafloor spreading history, the positive correlation between convergence rate and fractional peridotite—the fraction of subducting crust that is comprised of RAP—(and, by extension,



**Figure 4.** Modeled subducted residual abyssal peridotite over the last 5 Ma. Inserts show close-ups of some subduction zones discussed further in text. Holocene volcanoes are plotted on top from the Global Volcanism Project (2023). Global results of other parameters (e.g., fractional peridotite content, depth, dip etc. are available in Supporting Information S1). Results use a representative  $H_2$  production value of  $200 \text{ g per m}^3$  of reactant.

hydrogen production) simply occurs because a high convergence rate increases the flux of seafloor into a subduction zone. Thus, there is directly more volume of ocean crust (either RAP or not) available to react. Hence, many of the subduction zones ringing the Pacific Ocean are overall more fertile than the slow Calabrian subduction zone, despite the fact that Calabria has a higher fraction of RAP being subducted (Figures 4 and 5). This correlation is likely slightly skewed for two reasons in our analysis. First, as we only consider the last 5 Ma, subduction zones with fast convergence rates are able to fully occupy the maximum pressure of the antigorite stability field. If our analysis was extended further back in time, the correlation may weaken slightly, as subduction zones with slower convergence rates would become fully populated. Second, the majority of present-day subduction occurs within the Pacific Ocean, while RAP at present-day is predominantly located within the Atlantic, Arctic and Indian ocean basins (Merdith et al., 2019). When these three ocean basins close in the future, we would expect a much higher RAP flux into subduction zones (e.g., Grevemeyer et al., 2018). Further back in Earth history, we hypothesize that during times of supercontinent tenure and breakup (e.g., 320–180 Ma for Pangea) there is a reduced flux of RAP being subducted; however, when supercontinents assemble and internal ocean basins close (e.g., Rheic, Iapetus Oceans, future Atlantic/Indian Ocean) we speculate that there will be an increased flux,



**Figure 5.** Modeled H<sub>2</sub> production through “slab-serpentinization” of residual abyssal peridotite over the last 5 Ma. The conversion to hydrogen includes all points currently subducted, including those that have passed through the antigorite stability field, and those that are yet to have passed through. Inserts show close-ups of some subduction zones discussed further in text. Holocene volcanoes are plotted on top from the Global Volcanism Project (2023). Global results of other parameters (e.g., fractional peridotite content, depth, dip etc. are available in Supporting Information S1). Results use a representative H<sub>2</sub> production value of 200 g per m<sup>3</sup> of reactant.

possibly up to several orders of magnitude on the global scale (e.g., Grevemeyer et al., 2018; Merdith et al., 2019) s, depending on the configuration of subduction zones. This hypothesis is contrary to hydrogen production at mid-ocean ridges, which is greatest as a supercontinent breaks-up (Merdith et al., 2020).

#### 4.2. Viability of Our Global Estimate

An increasing number of field-based, experimental, and theoretical studies have shown the possibility for H<sub>2</sub>-rich, strongly reduced fluids to form through serpentinization reactions in subduction zones (Ferrando et al., 2010; Sachan et al., 2007; Vitale Brovarone et al., 2017, 2020). However, global fluxes of deep H<sub>2</sub> related to this process are largely unconstrained. One way to assess the viability of our global estimate is to compare our volumetric fluxes to other similar fluxes within ocean basins, such as gross crustal production rates and H<sub>2</sub> fluxes within different oceanic environments.



Gross oceanic fluxes, including subduction flux and mid-ocean ridge flux, have been estimated in previous studies (e.g., East et al., 2019; Jarrard, 2003; Zahirovic et al., 2022). Our calculated volumetric flux of all subducted RAP within subduction zones ( $1.84 \bullet 10^6 \text{ km}^3$  per Ma) represents  $\sim 10\%$  of the total volume of subducted oceanic crust over the same time range ( $1.5 \bullet 10^7 \text{ km}^3$ , East et al., 2019). Our volume flux of subducted RAP is slightly lower than estimates for the total volumetric exhumation of the mantle at ridges (8%–14%, Worman et al., 2016) because the average spreading rate at present-day ridges is lower than the average spreading rate at the time of the creation of the crust currently being subducted.

There are few studies that look at serpentinization of RAP within subducting slabs, limiting our ability to make direct comparisons of our calculated  $\text{H}_2$  flux ( $4.2\text{--}24 \bullet 10^7 \text{ kg/a}$ ). One prior estimate by Vitale Brovarone et al., 2017 suggested that a minimum of 0.8 Mt/a (or  $8 \bullet 10^8 \text{ kg/a}$ ) of  $\text{H}_2$  could be generated globally through the serpentinization of  $0.21 \text{ km}^3/\text{a}$  of RAP in subduction zones. Their calculation was done using the present-day exhumation rate of peridotite at mid-ocean ridges as a proxy for equivalent subduction rates, and used a generalized stoichiometric reaction of pure olivine and water to serpentinite, brucite, magnetite and hydrogen after Berndt et al. (1996) and McCollom and Seewald (2001). If we use the same equation as Vitale Brovarone et al. (2017), but substitute our volumetric estimates of total subducted RAP over the last 5 Ma (averaging to be  $1.8 \text{ km}^3$  per annum), we calculate  $\sim 6.8 \text{ Mt/a}$  (or  $6.8 \bullet 10^9 \text{ kg/a}$ ) of  $\text{H}_2$  being formed. For both examples, the amount of hydrogen generated is greater than the approach using the data of Vitale Brovarone et al. (2020) described in our results earlier. This is because the balanced equation used by Vitale Brovarone et al. (2017) does not consider the changes in pressure-temperature and fluid composition that occur in a subduction zone.

Other environments where serpentinization occurs include mid-ocean ridges and the forearc mantle wedge (Hyndman & Peacock, 2003; Williams & Gubbins, 2019). Our calculated  $\text{H}_2$  flux over the past 5 Ma ( $4.2\text{--}24 \bullet 10^7 \text{ kg/a}$ ) is 3–4 orders of magnitude smaller than what is estimated to occur at present-day mid-ocean ridges (MOR,  $\sim 10^{11}\text{--}10^{12} \text{ kg/a}$ , see Figure 8 in Merdith et al. (2020) for a comparison of  $\text{H}_2$  values from MOR environments), but is at the upper end of a previous estimated range for  $\text{H}_2$  generation through serpentinization of the forearc mantle wedge, of  $9 \bullet 10^6$  to  $3 \bullet 10^8 \text{ kg/a}$  (Vitale Brovarone et al., 2020). If we follow the logic of Vitale Brovarone et al. (2020), who calculated their flux by water availability in the downgoing slab and assume that all water stored in the RAP-derived antigorite ( $\sim 15 \text{ wt}\%$ , and all RAP above the minimum stability of antigorite) from our study is liberated into the forearc mantle wedge, we estimate between  $3.4 \bullet 10^5$  and  $1.8 \bullet 10^7 \text{ kg/a}$  of  $\text{H}_2$  could be produced from mantle wedge serpentinization (one order of magnitude less).

Though not explicitly considered in our model, the deep hydration of the mantle lithosphere during subduction due to trench flexure (Ranero et al., 2003) is thought to induce extensive serpentinisation, with maximum estimates of  $\sim 30\%$ – $40\%$  serpentinisation (3–5 wt% hydration) of the first 30 km of lithospheric mantle (Carlson & Miller, 2003; Van Avendonk & Holbrook, 2011). Based on results from thermodynamic modelling and geophysical constraints, Faccenda (2014) proposed a more conservative estimate whereby on average 10% of the upper 5–10 km of the lithospheric mantle may be serpentinised ( $\sim 1.2 \text{ wt}\% \text{ H}_2\text{O}$ ). Using our gross subduction flux and assuming a hydrated upper mantle after Faccenda (2014) equates to a peridotite flux of  $13.5\text{--}27 \text{ km}^3$  per annum reacting with (sufficient) water to produce  $\sim 2.7\text{--}5.3 \bullet 10^9 \text{ kg}$  per annum of  $\text{H}_2$  (1–2 orders of magnitude greater than our estimate). There is, however, some large ambiguity about exactly how well hydrated the upper mantle is.

The existence of deep-faults from trench flexure is well established from seismic observation (e.g., Han et al., 2016; Ranero et al., 2003) and the resulting alteration is inferred from variations in seismic velocities (e.g., Carlson & Miller, 2003), but interconnecting alteration fronts between faults (into unfractured mantle) have been questioned based on rheological and seismic grounds (Hatakeyama et al., 2017; Korenaga, 2017; Miller et al., 2021). For instance, the Middle American Trench (MAT, outboard of Nicaragua) is thought to have the most hydrated mantle (due to trench flexure) of all subduction zones (e.g., Grevemeyer et al., 2018). Miller et al. (2021) proposed that the anisotropy observed within the trench rise of this subduction zone is best explained by discrete serpentinisation fronts, focussed around faults, without an interconnected reaction front. They suggest that at this subduction zone the bulk degree of serpentinisation and the corresponding hydration rates of the upper mantle (up to 10 km depth) are 6.1%–8.8%, corresponding to 0.60–0.87 wt%  $\text{H}_2\text{O}$ , respectively (Miller et al., 2021; Miller & Lizarralde, 2016). This estimate is roughly half that of Faccenda's (2014) global estimate and ca. 5–10 times our estimate from RAP, but, if propagated globally, would likely slightly over-estimate the budget due to the more abundant hydration at the MAT relative to other subduction zones.

Our estimates of volumetric peridotite subduction are a similar order of magnitude, but slightly lower than what is produced at mid-ocean ridges, which makes conceptual sense given that the majority of slow and ultraslow spreading ridges currently occur in ocean basins without extensive subduction zones (Merdith et al., 2020, e.g., 2019). A clear implication of this is that once the Atlantic and Arctic oceans begin closing, there is likely to be a much larger flux of RAP into global subduction systems (Grevemeyer et al., 2018). Our results are roughly equivalent (in terms of peridotite that reacts to form serpentinite and hydrogen) to the lower bounds of Faccenda (2014), or the upper bounds if the conclusions of Miller et al. (2021) are used. These results suggest that, globally, there could be a similar amount of RAP being subducted and serpentinitised as what is expected to occur due to trench flexure and upper-mantle hydration.

### 4.3. Ground-Truthing Our Model

Due to limitations in acquiring in situ measurements of active subduction zones, evaluation or ground truthing of our model is limited to comparing evidence of reactants (i.e., presence of RAP in a subduction zone *indicating possibility of slab-serpentinization*) or products (slab serpentinitization, hydrogen generation) in areas where our model suggests slab-serpentinization is occurring. Unfortunately, many of these comparisons are indirect and hampered by arc processes, the thick over-riding plate that alter original signals, biosphere, or through our inability to distinguish slab-serpentinization signatures from abyssal serpentinite, making it difficult to draw quantitative conclusions (Martin et al., 2020). Instead, methodological field-work and detailed petrography remain some of the best tools for evaluating slab-serpentinization in exhumed terranes (e.g., Vitale Brovarone et al., 2021) and consequently we infer our correlations as a conservative upper bound for slab serpentinitization.

The slab of the lesser Antilles subduction zone is Cretaceous in age, that was originally formed at an ultraslow-spreading ridge (Seton et al., 2012) and is currently converging at around 20 mm/a with a dip angle of 30°. IODP cruises that dredged and cored the ocean crust outboard of the trench have returned serpentinitized abyssal peridotite and residual abyssal peridotite, indicating that slow-spreading ocean crust is likely a major component of the oceanic crust and the slab (Klein et al., 2017). Seismic analysis of the Lesser Antilles slab suggests the presence of hydrated slow-spreading crust penetrating deeply (100 km) into the subduction zone (Bie et al., 2022; Paulatto et al., 2017). While a large and hydrated sediment budget may also account for the seismic anomalies, Paulatto et al. (2017) argue that the enrichment of isotopically heavy Mg in the lavas of Martinique Island is best explained by serpentinite-derived fluids from the subducting slab. Hick et al. (2022) came to a similar conclusion using seismic attenuation models, showing that the Lesser Antilles Island Arcs were likely formed through devolatilization of the subducting slab, with the pre-subduction tectonic history of the ocean slab imparting a strong control on the evolution of the mantle wedge and arc. Above the forearc mantle, fluid inclusions from the volcanic arc of the Lesser Antilles preserve strongly positive  $\delta^{11}\text{B}$  signatures, interpreted to be derived from the serpentinitization of the oceanic mantle (Cooper et al., 2020).

Though our model, in this subduction zone, only represents the upper 20–50 km of subducted slab in this region, the earlier subducting seafloor (i.e., subducted prior to 5 Ma) is likely also enriched to a similar degree in RAP (e.g., Merdith et al. 2019). We speculate that the upper 7 km of our modeled slab and the deeper slab both are comprised of roughly 15%–20% of RAP, indicating that an extensive serpentinitization front close to the slab-forearc contact is possible. Assessment of the veracity of this hypothesis will depend on the source of fluids that cause serpentinitization in the RAP; if they are derived metamorphically then they are unlikely to produce such a positive  $\delta^{11}\text{B}$  signature; however, if the fluids are sourced through, for example, further hydration during trench flexure, then they could produce a similar  $\delta^{11}\text{B}$  signature.

In addition to seismic imaging of slabs, ophiolites in active subduction zones may preserve either direct or indirect signatures substantiating slab-serpentinization and/or an underlying ultramafic subsurface. Direct evidence of slab-serpentinization has been presented for some ophiolites through the European Alps and Appalachians (Boutier et al., 2021; Scambelluri & Tonarini, 2012; Vitale Brovarone et al., 2020, 2021), a region our model suggests is rich in the subduction of RAP. In these ophiolites, a strongly positive  $\delta^{11}\text{B}$  signature is interpreted to be indicative of serpentinitization by seawater, rather than derived from the progressive dehydration of subducting oceanic crust, thus supporting serpentinitization of the downgoing slab (e.g., Scambelluri & Tonarini, 2012).

The Philippines archipelago straddles a complex set of tectonically dissected microplates sandwiched between the Manila and Philippine Trenches. The subsequent Zambales ophiolitic section, west from the Luzon Fault,

emplaced in the Oligocene to early Miocene, is famous for springs, several of which exhibit geochemical patterns indicative of active serpentinization in the ultramafic subsurface, rich in  $H_2$  and/or  $CH_4$  (Cardace et al., 2015). Though slightly older than the time region of analysis for our model, but probably representing a slightly older portion of the Philippines seafloor which subduction aborted, our results support the possible emplacement of extensive ultramafic bodies in this region, with the Philippine trench represented in our model much more abundant in RAP and subsequent  $H_2$  than other areas (e.g., Figures 3a and 3b). The nearest neighbor taxonomic affiliations of the microorganisms sampled in the springs obtained from  $^{16}S$  sequences show that the microbial communities are dominated by hydrogen oxidizers and archaeal methanogens genera. While not direct evidence of slab-serpentinization, the emplacement and subsequent serpentinization of residual abyssal peridotitic bodies in this region support our central proposition that RAP is subducting and could possibly undergo serpentinization in the slab.

#### 4.4. Implications for Subduction Systems and Hydrogen Generation

Our results have several broad implications for the nature of subduction systems and the generation of abiotic hydrogen (and methane). However, we stress that our model, while novel, is coarser than regional seismic tomography models and is not numerically coupled with either fluid, geodynamic, or petrological models depicting subduction zone processes (though this could be a future possibility). Here, we discuss our results with reference to general hypotheses about slab serpentinization processes.

Oceanic cold-deep seeps on active subduction zones are places where emissions of ambient-temperature fluids with high concentrations of  $CH_4$  seep from the lithosphere into the hydrosphere (Torres & Bohrmann, 2016). Many (but not all) of these seeps are found on margins poor in organic-rich sediments. For example, the compilation of cold-deep seeps offered by Torres and Bohrmann (2016) and also Suess (2014) show cold-deep seeps laying on both RAP-rich subduction zones (from our analysis, such as the European Alps down toward Turkey, the Antilles and Sumatra) and the organic carbon-poor subduction zones of Plank and Manning (2019), such as Cascadia and the Aleutians. In the absence of a clear organic origin, extensive serpentinization events leading to the generation of  $CH_4$  plumes could sustain these seeps. In particular, either serpentinized RAP (e.g., Meister et al., 2018; Sciarra et al., 2019) or where fracture zones subduct, allowing localized hydration of the upper mantle, could be a viable source of these seeps.

Causes of volcanic and arc gaps, segments of a continental arc that do not produce active volcanism despite ongoing evidence of subduction (McGeary et al., 1985), remain enigmatic. At present-day, the most predominant volcanic gaps occur along the South American Andean margin (Ramos, 1999) and in the Aleutians (Eberhart-Phillips and Christensen, 2006; the Denali Gap, e.g., Plafker & Berg, 1994). As continental arc volcanism is driven predominantly by partial melting in the mantle wedge and subducting slab (Gianni & Luján, 2021; Grove et al., 2012; Peacock, 1990), hypotheses to account for the absence of volcanism predominantly pertain to a combination of tectonic factors that essentially limit fluid generation. The most prominent hypotheses being flat slab subduction (dip angle  $<30^\circ$ , typically thought to occur because of slab buoyancy), which inhibits melt generation because the slab is unable to warm up sufficiently to induce melt, or thickened over-riding crust, where the mantle wedge remains cold and also inhibits the geotherm. Serpentinization, especially of RAP or the lithosphere in locations of thinned crust, are a possible intriguing piece in this puzzle, as they directly relate to processes that increase melt (due to abundance of water, possibly increasing volcanism (Grove et al., 2012)) and inhibit melt (through buoyancy, decreasing melt (Yang et al., 2020)).

To first order, our results indicate that serpentinization of the RAP is more strongly correlated with volcanism than with volcanic gaps (e.g., Figure 3, though we cannot omit that this could also correlate with serpentinite rich, slow-subducting lithosphere). This correlation is clearest in the Andean margin, where the one region with enriched RAP relative to the rest of the margin is beneath southern Peru and northern Chile, where there is abundant volcanism. In the northern volcanic gap along the Andean margin, our model suggests that a much smaller amount of RAP is likely being subducted (Figure 3). Along the Aleutian subduction zone, a similar transition is also observed, where low amounts of RAP are subducted in the NE corner of the subduction zone, where the Denali Gap occurs. However, these sections (Andean margin, Aleutians), while enriched relative to the rest of their margins, still contain far less RAP than other margins (Figure 4). Our overall results suggest that there could be some threshold in the flux of subducting RAP content that plays an important role in controlling the geometry and tectonic parameters of a subducting slab.

## 5. Implications and Conclusions

Recent studies have begun highlighting the nuance required to untangle subduction zone processes, in particular distinguishing the effect of localized slab features (e.g., fracture zones, thinned crust) and a heterogeneous slab composition on fluid generation and rheology of both the downgoing and overriding plate. Our analysis here couples discrete data sets, concretely linking the seafloor spreading history of present-day ocean basins to estimate the global potential of “slab-serpentinization” and resulting hydrogen generation. In particular, our results broadly support recent observations that reduced fluids can be generated in subduction zones (Vitale Brovarone et al., 2020), which have also been documented in the shallow forearc (e.g., Debret et al., 2022; Plümper et al., 2017). The framework we have constructed as part of this analysis can also be used to test different hypotheses about transport and interaction of volatiles and rocks from ocean basins into subduction zone settings (e.g., formation of magnetite as a product of slab-serpentinization and implications of deeper H<sub>2</sub> generation, transport of carbon, inter-relationships between slab age and subduction processes).

We identify two key controls on H<sub>2</sub> production within the serpentinite window in subducting slabs. First, H<sub>2</sub> production is dependent on determining how much raw material is available to undergo serpentinization. Our analysis identifies seafloor spreading rate at the time of crust formation as being the strongest tectonic control on possible H<sub>2</sub> production. Second, the P-T conditions of a subduction zone control the limit of the antigorite stability zone. The approach we have followed here allows us to estimate a minimum amount of hydrogen generated from subducting slabs, using volumes of RAP (connected directly to the seafloor spreading history of ocean basins) entering a subduction zone and determining the mass of RAP that is currently within, or has passed through, the antigorite stability zone. Our results suggest that up to 10<sup>8</sup> kg/a of hydrogen could be generated through slab-serpentinization across global subduction zones (3–4 orders of magnitude lower than at mid-ocean ridges), and we suggest that some well-identified volcanic gaps correlate with regions of less RAP being subducted.

## Data Availability Statement

All data and code used for this analysis are available at Merdith et al. (2023), as well as at [github.com/amer7632/Merdith2023\\_pgpSlabs\\_G3](https://github.com/amer7632/Merdith2023_pgpSlabs_G3) (where future updates will be stored).

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