Introduction to the used geochemical agents

Our database contains an extensive collection of isotope and corresponding elements (ratio) data, showing strong correlations with dehydration and redox cycles in previous studies (see below for references). We have mapped the geochemical data using geographic coordinates. This effectively distinguishes between arc and back-arc samples and clarifies various trends. Although some agents may originate from different rocks, they can exhibit similar trends. Some agents in the same sample also showed coupling relationships (e.g., $\delta^{56}Fe$ and B/Nb; H_2O and Fe^{3+}/Fe_{tot}), suggesting coupling relationships between different isotope systems from arc to back-arc systems. In addition, we present the average values of the compiled data in this study and different subducting rock layers from previous studies (Figure 4 and Table S3). This allows readers to easily reference relevant values. This database plays a crucial role in validating our model. The accompanying document provides detailed principles and related references for using these data, supporting the descriptions in the main text. It is important to note that geochemical analyses often yield multiple interpretations, and some proxies are influenced by more than one factor. However, if our model successfully explains all the related proxies, it represents a highly robust mechanism.

Sulfur isotope system and sulfur contents

The processes of partial melting, sulfide segregation, and degassing could potentially contribute to the enrichment of $\delta^{34}S$ in arc volcanic rocks. However, due to the possibility of sulfur-undersaturated melting and the lack of corroborative data, these processes are unlikely to be the primary drivers. Instead, the $\delta^{34}S$ values in arc volcanic rocks likely reflect the sulfur composition of their mantle sources (Alt et al., 1993). It is probable that the sulfur in Mariana arc lavas originates from a depleted mantle source region that has been metasomatized by subduction-zone fluids. Alt et al. (1993) proposed that these sulfate-rich fluids are primarily derived from sediment and basaltic oceanic crust. However, subsequent research has revised these conclusions. For example, the mean $\delta^{34}S$ of altered oceanic basement only slightly deviates from the protolith, suggesting that the subduction of altered basaltic crust is unlikely to be the source of the high $\delta^{34}S$ values observed in arc volcanic rocks (Alt et al., 1995).

Recent datasets (Li et al., 2020) provided further evidence, showing that both subducting sediments and basaltic rocks exhibit negative sulfur isotope ratios (Table S3). In addition, mass balance calculations have ruled out subducting sediments and altered oceanic crust (AOC) as significant sources of elevated sulfur isotopes in arc magmas, given their thin layers. In contrast, only serpentinized rocks (i.e., partially hydrated mantle, as referenced in this study) exhibit high isotope ratios, reaching values up to +20‰ (Li et al., 2020). Recent studies (Schwarzenbach et al., 2024) have demonstrated that serpentinized rocks can release heavy sulfur isotopes in subduction zones, providing a plausible explanation for tracing the sulfur sources in arc magmas.

In our database, the Mariana arc magma displays heavy sulfur isotope ratios (average value=6.79‰) ranging from +0.1‰ to +20.7‰ (Fig. 4 and Table S2), which align with the isotope signatures observed in serpentinized rocks (Li et al., 2020). Previous studies have indicated that the sulfur content in Mariana arc magma is higher than that in MORB (1500 ppm), averaging 2000 ppm (Brounce et al., 2015, 2019). These sulfur isotopes and contents data suggest that partially hydrated mantle contributes a substantial amount of sulfur to the sub-arc mantle through fluid-driven desulfurization. In contrast, back-arc magma exhibits sulfur isotope values and average sulfur content similar to those of MORB (Fig. 4, Brounce et al., 2019), implying that sulfur-rich fluids are depleted at back-arc depths and are not responsible for the oxidation mechanisms in back-arc magma. Our sulfur-related database, in conjunction with previous research, supports our proposed mechanism and validates the accuracy of mass balance calculations.

Boron isotope system and B/Nb ratio

Boron isotopes are a powerful tool for tracing the presence of serpentinite-derived fluids (e.g., Zhang et al., 2021; Marschall et al., 2018). The upper mantle is typically characterized by a depletion of $\delta^{11}B$, with boron concentrations of less than 0.1 ppm and $\delta^{11}B$ values ranging from -10‰ to -7‰. In contrast, serpentinized rocks exhibit significant enrichment, with boron concentrations reaching up to 100 ppm and $\delta^{11}B$ values as high as +41‰ (Zhang et al., 2021). Other rock layers generally have lower boron isotope values (Table S3). The high $\delta^{11}B$ values in serpentinized rocks are attributed to serpentinization by seawater-derived fluids that have interacted with mantle rocks prior to the serpentinization process, resulting in the strong enrichment of isotopically heavy boron (Marschall et al., 2018).

The B/Nb ratio, particularly elevated in serpentinized rocks, serves as an additional proxy for identifying subducted serpentinite and has been widely used in previous studies (e.g., Zhang et al., 2021; Chen et al., 2023). This ratio has been confirmed to be unaffected by magmatic processes (Ryan et al., 1996; Ottolini et al., 2009), as boron behaves as a highly incompatible element during these processes (i.e., partial melting and crystallization differentiation). When boron is compared to other highly incompatible elements, such as niobium (Nb), the ratio forms a compact array passing through the origin, indicating a constant ratio of incompatible elements that reflects the characteristics of the mantle source (Ryan et al., 1996). Consequently, the observed positive correlation between $\delta^{11}B$ values and B/Nb ratios in arc lavas is interpreted as evidence of serpentinite fluid contributions to the subarc mantle wedge.

In our boron-related database, the $\delta^{11}B$ isotope range in arc magmas is +2.86‰ to +6.2‰, with an average value of +4.63‰, which is significantly higher than that of MORB (-7.1‰, Table S3). This suggests deep penetration of high-B fluids and underscores the contribution of serpentinized rock (±AOC). The B/Nb ratio follows the same trend, with an average value of 1.2 for MORB (Table S3), which is lower than the arc B/Nb ratio in our database (10.58, Table S2). In contrast, the B isotopic values and B/Nb ratios of the back-arc samples (-4.74‰ and 3.28, respectively; Table S2) are closer to those of MORB, indicating a weaker influence of fluids. This also suggests that slab-derived fluids are not responsible for the oxidation of back-arc magma.

Chlorine isotope system and Cl/Nb ratio

Chlorine isotopes are another powerful tool for tracking plate dehydration and contributions from different rock layers, and they have been extensively used in the Mariana system due to strict controls from different values come from different subducting rock layers (Barnes et al., 2008, 2010). In general, oceanic sediments have negative $\delta^{37}Cl$ isotope ratios, mostly ranging from -2.0‰ to 0‰, with an average of -1.1‰ (Barnes et al., 2008). Altered oceanic crust also shows negative $\delta^{37}Cl$ values, with an average of -1.3‰. The $\delta^{37}Cl$ value of

MORB is reported to be 0‰, while serpentinized rock typically has a high average $\delta^{37}Cl$ of +0.2‰ (Fig. 4; Barnes et al., 2008).

Previous studies on the Mariana front arc (<130 km depth) indicate that it typically has $\delta^{37}Cl$ value, falling within the isotopic range of altered oceanic crust and sediments (Fig. 4; Barnes et al., 2008), which suggests contributions from sediment and AOC. However, analysis of the rear arc (130−230 km depth) reveals a positive value of +0.2‰ (Fig. 4), and this can only be attributed to the dehydration of serpentinized rocks (Barnes et al., 2008). This finding aligns with our understanding of the dehydration process (Fig. 1). In contrast, rocks from back-arc area exhibit negative $\delta^{37}Cl$ values (-0.76‰), indicating that dehydration has been completed at this stage, but the contribution of sediments and AOC by partial melting (Fig. 2), potentially attributed to the instability of white-mica as depicted in the CFE models.

The Cl/Nb ratio is also often used as a marker for dehydration (e.g., Barnes et al., 2010; Sun et al., 2007), also because niobium is usually very stable as an incompatible element in fluid. When chlorine levels increase in the arc-related rocks because of fluid, the Cl/Nb ratio also increases. It is estimated that ~80% of the Cl in the Manus arc-type glasses was added directly from subducted slab-derived fluids (Sun et al., 2007), resulting in an increase in the Cl/ Nb ratio.

It is important to note that to minimize the impact of degassing, all Cl/Nb (/100) data are derived from melt inclusions in olivine (Table S2). Compared to MORB and back-arc samples (0.21 and 3.87, respectively), the Cl/Nb/100 ratio in arc samples is significantly higher, with an average of 12.02 (Fig. 4), indicating that a substantial amount of chlorine entered the arc magma via fluids.

Copper isotope system and copper content

Copper is a chalcophile element that is highly sensitive to redox conditions (Lee et al., 2012; Wang et al., 2018). During magma differentiation, sulfur saturation and the segregation of sulfides can lead to copper depletion and potentially cause an enrichment of heavy copper isotopes in the residual magma. In contrast, in sulfide-undersaturated evolved magmas, copper tends to become enriched, with minimal fractionation of copper isotopes (e.g., Huang et al., 2012). During partial melting in the mantle, copper behaves as an incompatible element, showing little isotope fractionation (Lee et al., 2012). However, the presence of oxidative fluids produced by the dehydration of subducting plates and mantle metasomatism can cause significant fractionation of copper isotopes (Liu et al., 2015; Chen et al., 2022). This process is considered the primary factor contributing to the broad range of $\delta^{65}Cu$ values observed in metasomatized mantle peridotites and one of the reasons for the heavier copper isotopic composition found in arc-related lavas (Liu et al., 2015).

Moreover, the composition of fluids plays a critical role in determining copper isotope fractionation between minerals and fluids. $Cu^{2+}SO_x$ complexes in the fluid, for instance, are theoretically more likely to concentrate heavy copper isotopes (e.g., Fujii et al., 2014). Given strong affinity of copper for sulfur, it demonstrates high mobility via sulfur complexes during slab dehydration. Recent research has shown that the elevated $\delta^{34}S$ values observed in arc lavas further support the involvement of sulfate-rich oxidizing fluids in the subarc mantle metasomatism (Chen et al., 2022). Serpentinite also tends to have relatively high $\delta^{65}Cu$ values (average value=+0.42‰; Chen et al., 2022; Liu et al., 2015). Consequently, the sulfate-rich fluids released by subducted serpentinite at sub-arc depths are expected to carry a heavier isotopic composition of both copper and sulfur. As a result, the combined copper and sulfur isotopic signatures in arc-related lavas can serve as indicators of sulfate-rich fluids derived from serpentinite, which have metasomatized the sub-arc mantle.

In our database, the $\delta^{65}Cu$ isotopes in arc magmas (+0.51‰) are significantly heavier than those found in MORB (+0.06‰), and the copper content is also more enriched compared to MORB (105 and 77 ppm, respectively). This suggests that slab dehydration may be responsible for these observations, with the behavior of Cu reflecting the release of sulfate-rich oxidizing fluids at subarc area (e.g., Chen et al., 2022), consistent with our model results. In contrast, the $\delta^{65}Cu$ values and copper content in the back-arc magma (+0.14‰ and 61 ppm, respectively), which are similar to those of MORB, indicate that fluids did not contribute to the formation and oxidation of back-arc magma.

Iron isotope system and Fe^{3+}/Fe_{tot} ratio

Iron is a redox-sensitive element, and its isotopes can undergo fractionation during high-temperature geological processes (Dauphas et al., 2017; Weyer and Ionov, 2007). The fractionation of iron isotopes is strongly influenced by their redox states. Typically, trivalent iron (Fe^{3+}) is more incompatible than divalent iron (Fe^{2+}) during partial mantle melting, making Fe^{3+} more likely to enter the melt (e.g., Dauphas et al., 2009). Due to the stronger chemical bonds formed by Fe^{3+} compared to Fe^{2+}, Fe^{3+} tends to favor heavier iron isotopes, reflected in higher δ^{56}Fe values, while Fe^{2+} tends to favor lighter δ^{56}Fe isotopes (e.g., Sossi and Debret, 2021).

However, global oxidized arc magmas generally exhibit lighter δ^{56}Fe isotope compositions relative to mid-ocean ridge basalts (e.g., Chen et al., 2023; Williams et al., 2018). For instance, in our database, Mariana arc magmas have light δ^{56}Fe isotopes (+0.02‰), whereas back-arc and mid-ocean ridge rocks exhibit heavier and similar δ^{56}Fe values (+0.09‰ and +0.1‰, respectively). This paradox may be related to prior sub-arc mantle melt extraction or the infiltration of slab-derived fluids with light δ^{56}Fe isotopes into the sub-arc mantle (e.g., Chen et al., 2023).

Although direct simulation results are unavailable due to the absence of DEW-related species (Sverjensky et al., 2014), isotopic evidence suggests that iron in slab-derived fluids predominantly exists as Fe^{2+}, which is typically associated with lighter isotopes, as observed in serpentinized rocks (Debret et al., 2016; Chen et al., 2023). The dehydration of these serpentinite rocks not only releases fluids containing lighter δ^{56}Fe isotopes but may also transport oxidative sulfate complexes of $Fe^{2+}SO_x$ (± $Fe^{2+}Cl_2$; Debret et al., 2016; Chen et al., 2023), analogous to the form of copper complexation (Chen et al., 2022).

The sulfates in these fluids can oxidize iron in the mantle, converting Fe^{2+} to Fe^{3+}, thereby increasing the redox state of the mantle (Chen et al., 2023). This mechanism aligns well with our model, which proposes that the oxidation of the sub-arc mantle is driven by the desulfurization of subducting slabs, primarily composed of serpentinized rocks (Fig. 1). The compiled Fe^{3+}/Fe_{tot} ratio of inclusions (Fig. 4), as the most intuitive redox agent, demonstrated that the whole-rock Fe^{3+}/Fe_{tot} ratio of arc magma and back-arc magma are 0.24 and 0.18 respectively as compared to MORB (0.15).

Molybdenum isotope system and Ce/Mo ratio

Previous studies have employed molybdenum (Mo) isotope fractionation to investigate the redox conditions of fluids within the Earth's interior (e.g., Anbar, 2004). The fractionation of Mo isotopes is predominantly influenced by redox reactions, as Mo exhibits distinct chemical behaviors depending on its oxidation state, which leads to isotopic fractionation (Anbar, 2004; Willbold and Elliott, 2017). In oxidizing environments, Mo predominantly exists in higher oxidation states, such as Mo^{6+}, whereas in reducing environments, Mo can exist in lower oxidation states, such as Mo^{4+}. By measuring the isotopic composition of Mo, the redox state of the fluid can be inferred. Higher $\delta^{98/95}$Mo isotope values typically indicate strongly oxidizing conditions, while lower $\delta^{98/95}$Mo isotope values suggest that the fluid may be under more reduced conditions (König et al., 2016; Chen et al., 2019; Li et al., 2021).

Chen et al. (2019) explored the role of Mo isotopes as indicators of the redox state of fluids in subduction zones. Their findings demonstrate that Mo isotope fractionation is strongly linked to the oxidative nature of fluids present in high-pressure metamorphic rocks. These fluids are thought to originate from the dehydration of serpentinite in the mantle beneath subducting plates. During dehydration processes, heavier Mo isotopes preferentially partition into the fluid phase, resulting in a lighter Mo isotope composition in the associated metamorphic rocks. By analyzing Mo isotopes, we can trace the redox conditions and fluid migration pathways within subduction zones, providing insights into the geochemical processes occurring in these complex environments. Li et al. (2021) suggested that the heavy isotope characteristics in the Mariana arc magma are due to the subduction of mantle wedge serpentinite from forearc area. However, it is difficult to draw this conclusion solely from the Mo isotope characteristics. As mentioned before, the subducting lithospheric mantle can also release heavy Mo isotopes into the fluid. Additionally, the subduction of the forearc serpentinite is unlikely to transport water-bearing minerals to deep sub-arc regions (e.g., 150 km), as slab-top temperatures can easily dehydrate these serpentinites at shallower depths.

Another important redox indicator related to Mo is the Ce/Mo ratio. Under oxidizing conditions, Ce^{3+} is readily oxidized to Ce^{4+}, which is relatively insoluble (Trail, 2018). In contrast, Mo typically exists as Mo^{6+}, which is highly soluble and easily retained in fluids (e.g., Skora et al., 2017; Anbar, 2004). Consequently, in an oxidizing fluid, cerium is more likely to precipitate, while molybdenum remains dissolved, leading to a decrease in the Ce/Mo ratio. This ratio thus can reflect the redox state of the fluid. The observed low Ce/Mo ratio in arc magmas (e.g., Li et al., 2020) also suggests significant inputs of Mo, further highlighting the influence of redox conditions on Mo distribution (Chen et al., 2019).

In Marianas arc magma, $\delta^{98/95}$Mo isotope values are significantly heavier (-0.03‰) relative to those in MORB (-0.2‰) and back-arc magma (-0.31‰), indicating the addition of highly oxidizing fluids. According to prevailing theories, these fluids likely originate from the dehydration of serpentinized rocks. Additionally, the Ce/Mo value of the arc magma (=23) is relatively low, reflecting the oxidative nature of the fluid and the enrichment of Mo, likely due to Mo-rich fluid metasomatism (e.g., Chen et al., 2019).

Water content and Ba/La ratio

The water content in arc-related inclusions serves as a direct indicator of magma H_2O levels and offers an advantage over analyzing rocks, as it minimizes the impact of degassing. A particularly useful and commonly employed proxy for water content in magma is the Ba/La ratio (e.g., Kelley and Cottrell, 2009; Brounce et al., 2014). Ba is a highly fluid-mobile element, meaning it readily enters the aqueous phase during the dehydration of subducting slab. when the subducting plate dehydrates, Ba is released into the fluid and subsequently becomes enriched in the overlying sub-arc mantle through fluid transport. In contrast, La, a rare Earth element, is relatively immobile in fluids and tends to remain within minerals rather than entering the aqueous phase as easily as Ba does. Consequently, La is typically not significantly enriched in dehydrated fluids.

This difference in behavior results in a higher concentration of Ba in arc magma compared to La, leading to an increased Ba/La ratio (Kelley and Cottrell, 2009). This elevated ratio is a key indicator of fluid processes in subduction zones and provides insights into the water content and fluid dynamics of arc magmas.

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