

Constraints of boron and oxygen stable isotopes on dehydration fluids, sediment-derived melts, and crustal assimilation of the Toba volcanic system (Indonesia)

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ABSTRACT

Arc magmas are produced from the mantle wedge, with possible addition of fluids and melts derived from serpentinites and sediments in the subducting slab. Identification of various sources and their relevant contributions to such magmas is challenging; in particular, at continental arcs where crustal assimilation may overprint initial geochemical signatures. This study presents oxygen isotopic compositions of zoned olivine grains from post-caldera basalts and boron contents and isotopes of these basalts and glassy melt inclusions hosted in quartz and clinopyroxene of silicic tuffs in the Toba volcanic system, Indonesia. High-magnesian (≥ 87 mol% Fo [forsterite]) cores of olivine in the basalts have $\delta^{18}\text{O}$ values ranging from 5.12‰ to 6.14‰, indicating that the mantle source underneath Toba is variably enriched in ^{18}O . Olivine with < 87 mol% Fo has highly variable (4.8–7.2‰), but overall increased, $\delta^{18}\text{O}$ values, interpreted to reflect assimilation of high $\delta^{18}\text{O}$ crustal materials during fractional crystallization. Mass balance calculations constrain the overall volume of crustal assimilation for the basalts as $\leq 13\%$. The processes responsible for the ^{18}O -enriched basaltic melts are further constrained by boron data that indicate the addition of < 0.1 wt% fluids to the mantle, $> 40\%$ of the fluids being derived from serpentinites and others from altered oceanic crust and sediments. This amount of fluids can increase $\delta^{18}\text{O}$ of the magma by only $\sim 0.02\%$. Approximately 6–9% sediment-derived melt hybridization in the mantle wedge is further needed to yield basaltic melts with $\delta^{18}\text{O}$ values in equilibrium with those of the high-Fo olivine cores. The cogenetic silicic tuffs, on the other hand, seem to record a higher proportion of fluid addition dominated by sediment-derived fluids to the mantle source, in addition to crustal assimilation. Our reconnaissance study therefore demonstrates the application of combined B and O isotopes to differentiate between melts and fluids derived from serpentinites and sediments in the subducted slab—an application that can be applied to arc magmas worldwide.

INTRODUCTION

Subduction zones are vigorously dynamic plate-tectonic boundaries where dehydration fluids, sediment-derived melts, and oceanic crustal melts can be recycled through the mantle to the surface, yielding diverse chemical compositions

of arc volcanic rocks (e.g., Plank and Langmuir, 1993). Various geochemical indexes including trace element ratios (i.e., Ba/Th, La/Sm, $\text{H}_2\text{O}/\text{Ce}$) and radiogenic and stable isotopes (i.e., $^{87}\text{Sr}/^{86}\text{Sr}$, $\delta^{11}\text{B}$, $\delta^{98}\text{Mo}$) have been used to recognize components of the slab input in subduction zones (Elliott et al., 1997; König et al., 2016). However, due to the compositional similarity of the silicic continental crust to the mean of subducted sediments (Stern, 2002; Plank, 2014), the

ascent of arc magmas through a thick continental crust makes it difficult to disentangle mantle source characteristics from crustal assimilation. In particular, at the Toba volcanic system (Indonesia), one of the largest active continental arc magmatic systems with two super-eruptions in the Quaternary, the issue of source components of an enormous amount of silicic magmas ($> 8000 \text{ km}^3$) with high $^{87}\text{Sr}/^{86}\text{Sr}$ and low ϵ_{Nd} , compared to other volcanoes in the same arc, was revived due to the discovery of subducted terrigenous sediments with crustal Sr and Nd isotope compositions (Gao et al., 2022).

Our study, for the first time, uses a combination of boron isotopes of the most primitive basalts and oxygen isotopes of the olivine grains with forsterite (Fo) $\text{Mg\#}$ [$\text{Mg}/(\text{Mg} + \text{Fe}) \times 100$] values up to 89 mol% in the basalts to distinguish the contributions of fluids and melts derived from the subducted slab and crustal assimilation beneath the Toba volcanic system. Oxygen isotopes of olivine with different Fo values can constrain the processes of both fluids and melts added to the mantle wedge, as well as assimilation and fractional crystallization (AFC; Eiler et al., 2005; Deegan et al., 2021). The concentrations and isotopic compositions of boron can reflect the percentage of fluids derived from serpentinites or sediments at sub-arc depth (e.g., De Hoog and Savov, 2018; Cooper et al., 2020) and therefore can be used to deduce whether the amount of those fluids is sufficient to generate the observed oxygen isotope values. Finally, we demonstrate the usefulness of combined B and O isotopes in differentiating various components in complicated continental arc magmatic systems.

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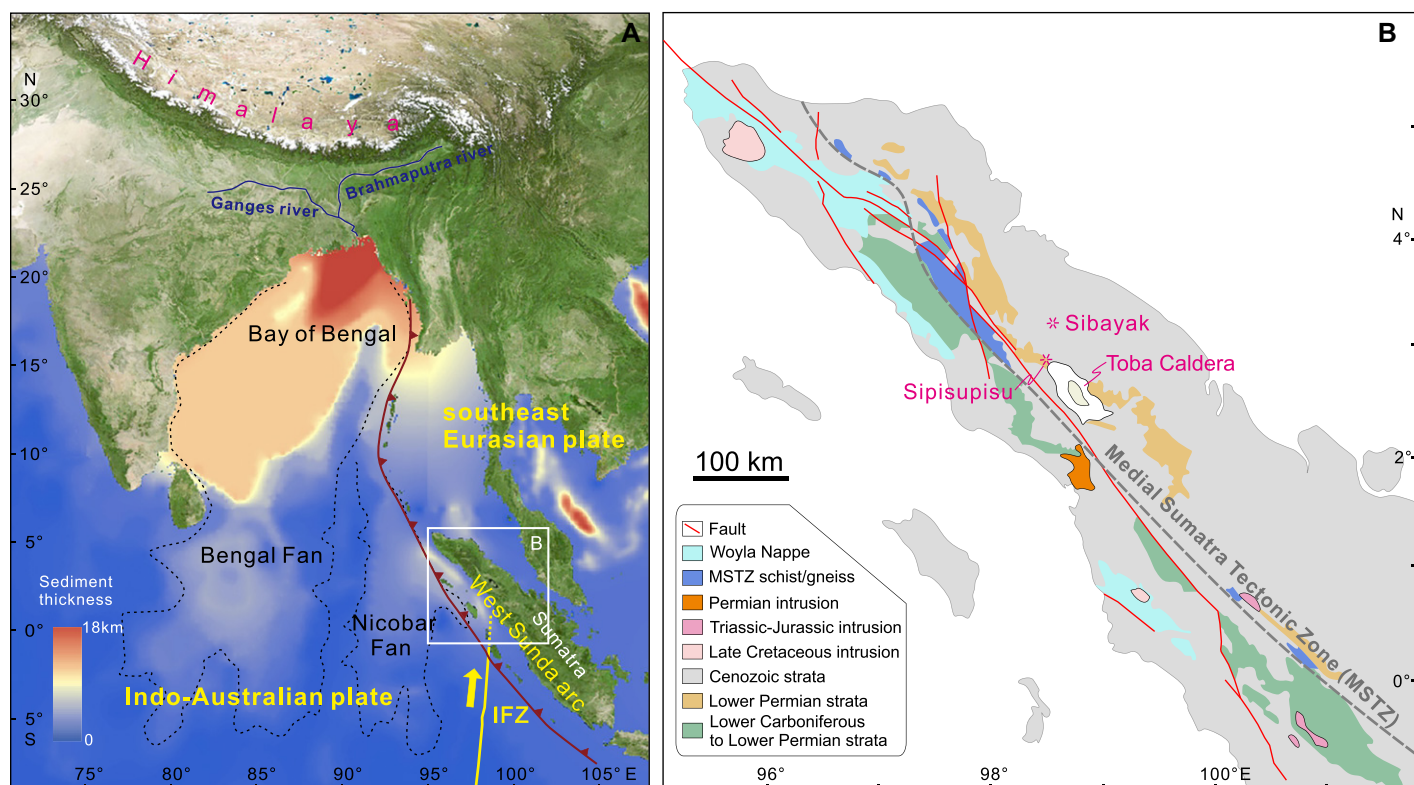


Figure 1. (A) Map of Southeast Asia. Sediment thickness is drawn after Straume et al. (2019). IFZ—Investigator Fracture Zone. (B) Map of northern Sumatra showing the Toba volcanic system.

THE SUMATRAN SUBDUCTION ZONE AND TOBA VOLCANIC SYSTEM

Sumatra is located at the convergent plate boundary between the Indo-Australian and southeast Eurasian plates (Fig. 1A). The Indo-Australian plate carries a thick sequence of terrigenous sediments (the Nicobar Fan) that are composed predominantly of sandy and muddy turbidites supplied by rivers from the Greater Himalaya and Gangdese arc and subducts north-northeast at an angle of $\sim 65^\circ$ below Sumatra (McNeill et al., 2017).

The Toba volcanic system in northern Sumatra (the west Sunda arc) is well known for producing four large silicic eruptions in the Quaternary, i.e., the ca. 1.42 Ma Harang-gaol Dacite tuff (HDT), the ca. 0.8 Ma Oldest Toba tuff (OTT), the ca. 0.5 Ma Middle Toba tuff (MTT), and the ca. 75 ka Youngest Toba tuff (YTT), together with several post-YTT lava domes and stratovolcanoes such as Sipisupisu volcano and Mount Sibayak (Fig. 1B). Among them, OTT and YTT were both super-eruptions. The footprint of the Toba magmatic system is considered to extend beyond Sipisupisu to Sibayak, which is ~ 40 km from the northern tip of the Toba Caldera (Mucek et al., 2017). The OTT, MTT, and YTT are typical silicic eruptions that are composed of dacite and rhyolite (Chesner, 1998). Quartz, sanidine, plagioclase, amphibole, and biotite are the main phenocrysts in these rocks. The HDT,

in contrast, is andesitic to dacitic and contains phenocrysts of clinopyroxene, plagioclase, and orthopyroxene. Basalts of Sipisupisu volcano represent the most mafic post-YTT volcanism and uniquely contain olivine phenocrysts, together with clinopyroxene, plagioclase, and orthopyroxene (Gao and Liu, 2023).

RESULTS AND DISCUSSION

Mantle Source Enrichment and Crustal Assimilation of the Toba Volcanic System

Long-lived volcanic systems associated with continental arcs are thought to be underlain by trans-crustal magmatic systems characterized by vertically stacked melt-rich lenses in the lower crust where melt segregation, country rock assimilation, and reactive flow may occur (e.g., Cashman et al., 2017). The Toba volcanic system produced episodic eruptions from ca. 1.5 Ma until at least 2.7 ka (Mucek et al., 2017), and the shallow magma reservoir feeding the two super-eruptions could have existed for up to 600 k.y. (Liu et al., 2021). Seismic tomographic studies indicate that a large basaltic magma reservoir is ponded at the Moho boundary underneath Toba, connected to the crustal magma reservoir by a large magmatic sill complex (Jaxybulatov et al., 2014). Because large volcanic systems are commonly dominated by silicic eruptions (Wilson et al., 2021), finding early crystallized minerals, such as olivine in equilibrium with partial melts of mantle perido-

rites, is the key to distinguishing mantle source enrichment from crustal assimilation.

The main eruptive phase of Toba comprised four silicic eruptions, where andesites of the HDT are the most primitive composition reported thus far and contain clinopyroxene with $Mg\# < 77$ and no olivine (Liu et al., 2022). However, basalts of the post-caldera Sipisupisu lava dome contain olivine grains with a high-Fo core (89–78 mol%) and a low-Fo rim (86–62 mol%) (Table S1 in the Supplemental Material¹). The olivine cores are typically 200–400 μm in diameter and unzoned (Figs. 2A–2C). The roughly linear correlations of Ni and Mn contents with Fo values indicate that the parental magma evolved via fractional crystallization of olivine (Figs. 3A and 3B). Notably, the three high-Fo (≥ 87 mol%) and high-Ni (> 1400 ppm) olivine cores have compositions close to those in equilibrium with mantle peridotite-derived melts (Fig. 3A), but with variable $\delta^{18}O$ values ranging from 5.12‰ to 6.14‰, mostly beyond the mantle range ($5.18\text{‰} \pm 0.28\text{‰}$; Matthey et al., 1994). Thus, we consider that the high Fo (≥ 87 mol%) olivine cores were crystallized from isotopically heterogeneous basaltic

¹Supplemental Material. Detailed description of the analytical methods, Figure S1, and Tables S1–S3. Please visit <https://doi.org/10.1130/GEOLOGY.S24760170> to access the supplemental material; contact editing@geosociety.org with any questions.

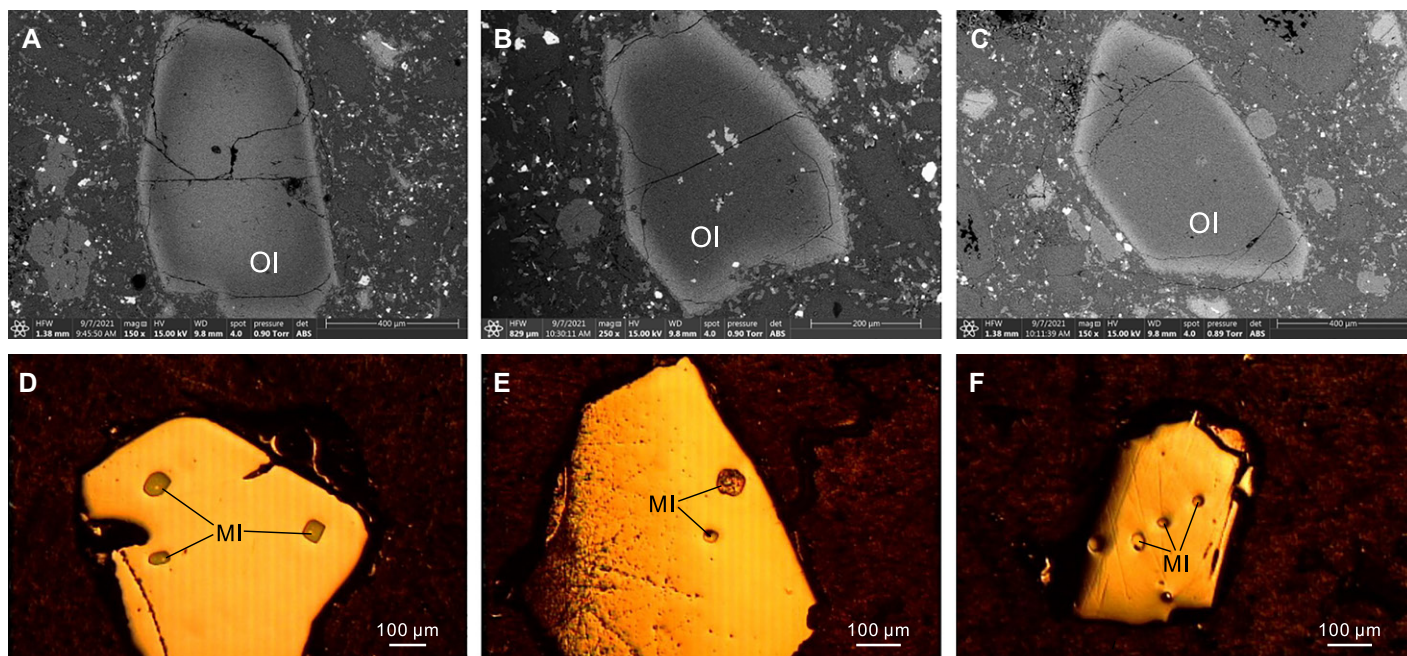


Figure 2. (A–C) Backscattered electron images of zoned olivine (OI) grains in the studied basalts from the Toba volcanic system, Indonesia. Glassy melt inclusions (MI) in quartz of the Youngest Toba tuff (YTT) and Oldest Toba tuff (OTT) eruptions (D, E) and in clinopyroxene of the Haranggaol Dacite tuff (HDT) eruption (F).

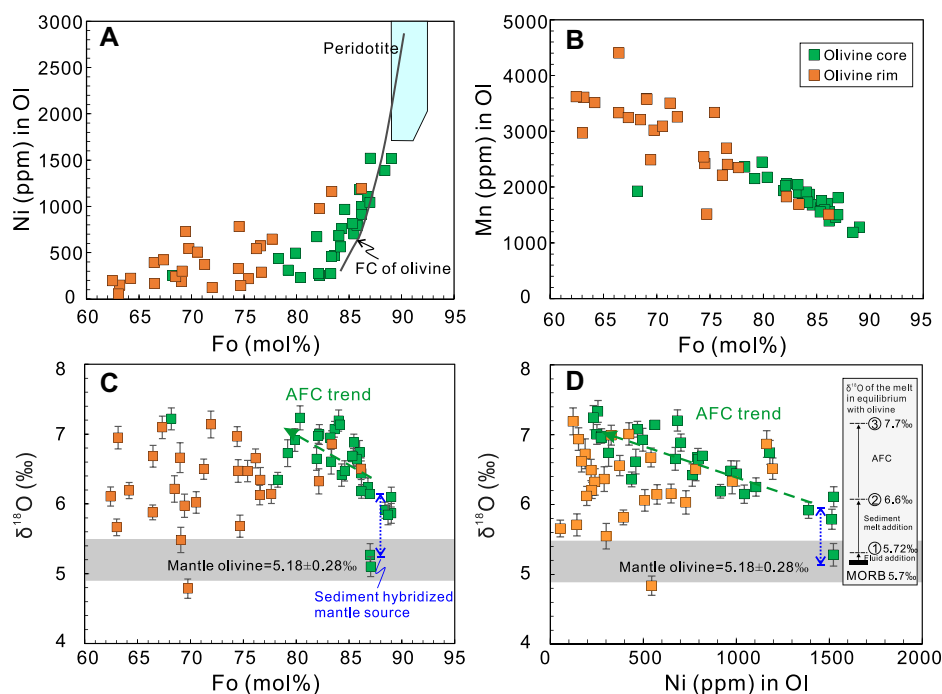


Figure 3. Geochemical plots for zoned olivine (OI) grains of the studied basalts from the Toba volcanic system, Indonesia. Plots of Ni content versus Fo (A) and Mn content versus Fo (B) display roughly linear trends dominated by fractional crystallization. The blue field indicates olivine compositions in equilibrium with peridotite-derived melts (see Straub et al., 2011). The 1σ errors of Ni and Mn contents vary from 4% to 6% and from 5% to 10%, respectively. Plots of $\delta^{18}\text{O}$ versus Fo (C) and $\delta^{18}\text{O}$ versus Ni content (D) exhibit assimilation and fractional crystallization (AFC) trends. Replicate analyses of $\delta^{18}\text{O}$ of olivine cores with Fo ≥ 87 mol% are also shown in C. Inset in D illustrates a change of $\delta^{18}\text{O}$ of the mantle-derived melts by $\oplus 0.1\%$ fluid addition, and $\oplus \sim 6\text{--}9\%$ sediment-derived melt addition, followed by $\oplus < 13\%$ crustal assimilation of high- $\delta^{18}\text{O}$ materials. MORB—mid-ocean ridge basalt.

melts, which may have the highest $\delta^{18}\text{O}$ values of 6.64‰ (in equilibrium with $\delta^{18}\text{O}_{\text{olivine}}$ of 6.14‰ ; Eiler, 2001) derived from partial melting of a variably ^{18}O -enriched mantle source. The slightly low Ni contents (1517 ppm) of the olivine cores with Fo of ~ 89 mol%, compared with those in equilibrium with partial melts of mantle peridotites, could be due to different partition coefficient of Ni in melts with different major element compositions (Wang and Gaetani, 2008). Combined with the crustal-like Sr and Nd isotopic compositions of basalts and silicic rocks of the Toba volcanic system (Liu et al., 2022), the mantle source underlying Toba could represent a sediment-rich end-member on the west Sunda arc, in contrast to the sediment-poor end-member with mantle-like Sr-Nd-Pb-O isotopic compositions underlying the east Sunda arc (Deegan et al., 2021).

On the other hand, $\delta^{18}\text{O}$ values of olivine with Fo from 87 mol% to 78 mol% increase with decreasing Fo and Ni contents (Figs. 3C and 3D), which indicates assimilation of high- $\delta^{18}\text{O}$ crustal materials during fractional crystallization. The O isotopic heterogeneity increases for olivine with Fo < 78 mol%, indicative of their growing in a large heterogeneous system with multiple assimilants or a variable extent of assimilation. If we assume the primary melts of the Toba volcanic system have compositions close to the average primitive calc-alkaline arc basalts worldwide, with 49–51 wt% SiO_2 (Schmidt and Jagoutz, 2017), and the crustal melts at 1 GPa underneath Toba have ≥ 68 wt%

SiO₂ (Petford and Gallagher, 2001), mass balance calculations indicate that the most primitive basalts (SiO₂ = 51.4 wt%) of the Toba volcanic system could be formed by <13% crustal assimilation. This, in turn, can place constraints on the $\delta^{18}\text{O}$ values of the assimilants. If the O isotope of the melt (7.7‰) is in equilibrium with olivine with the highest $\delta^{18}\text{O}$ value of 7.2‰, the mantle-derived basaltic melts with $\delta^{18}\text{O}$ values of 6.64‰ need to assimilate crustal materials with an average $\delta^{18}\text{O}$ value of ~15‰. This value is significantly higher than that of the average lower continental crust ($\delta^{18}\text{O}$ = 8.1‰; Simon and Lécuyer, 2005) and indicates that high- $\delta^{18}\text{O}$ crustal materials could exist underneath Toba. Therefore, recycling of subducted materials and crustal assimilation both play important roles in generating the Sr-Nd-Hf-O isotopic characteristics of the Toba volcanic system. Considering that significant lower crustal assimilation also occurred at the plume-related Snake River Plain–Yellowstone volcanic province (western United States; Savov et al., 2009), this phenomenon should be recognized in long-lived large magmatic systems.

Contributions of Fluids and Melts Derived from Serpentinites and Sediments to the Mantle Source Enrichment of the Toba Volcanic System

Fluids and melts derived from serpentinites and sediments are potential candidates with high $\delta^{18}\text{O}$ values that can be added to the mantle wedge and therefore increase the $\delta^{18}\text{O}$ values of mantle-derived melts in subduction zones (e.g., Bindeman et al., 2005). The calcu-

lated $\delta^{18}\text{O}$ values (up to 6.64‰) of the primitive basaltic melts of the Toba volcanic system are ~0.9‰ higher than those of mid-ocean ridge basalts ($5.7 \pm 0.2\text{‰}$; Ito et al., 1987). To further constrain whether subduction-zone fluids alone can increase the $\delta^{18}\text{O}$ values of melts by ~0.9‰, we analyzed B contents and isotopes of the post-caldera basalts in Sipisupisu volcano and Mount Sibayak. Boron isotopes and B/Nb ratios are used to study mantle source characteristics of magma because they do not vary with partial melting and fractional crystallization (Fig. S1; Ishikawa and Tera, 1997; Marschall et al., 2017).

$\delta^{11}\text{B}$ values of the basalts vary from -2.05‰ to 0.27‰ (Table S2), which are higher than those of the depleted mantle ($\delta^{11}\text{B}$ = -7.1‰ ; Marschall et al., 2017) and within the range of global arc volcanic rocks (De Hoog and Savov, 2018). Possible fluid/magma sources for the coupled variations of $\delta^{11}\text{B}$ and Nb/B include serpentinites scraped from forearc mantle and from the ultramafic part of the slab, altered oceanic crust (AOC), and sediments in the subducting slab and continental crust. Forearc serpentinite-derived fluids typically have higher $\delta^{11}\text{B}$ values than slab serpentinite-derived fluids (Tonarini et al., 2011). Our data, however, cannot distinguish whether the serpentinite fluids come from the subducting slab or recycled forearc mantle. Mixing calculations indicate that addition of ~0.05–0.1 wt% fluids to the source of the magma, with >40% derived from serpentinites and <60% from AOC and sediments, may account for the B compositions of the basaltic melts (Fig. 4). If this amount of fluids were

added to the mantle-derived melts with an average $\delta^{18}\text{O}$ value of 5.7‰, calculation shows that the $\delta^{18}\text{O}$ values of the melt would only increase to ~5.72‰, significantly lower than that of the primary melts of Toba ($\delta^{18}\text{O}$ = ~6.6‰; inset in Fig. 3D). Therefore, we infer that sediment-derived melts were also added to the mantle source underneath Toba. If we assume that $\delta^{18}\text{O}$ values of the subducted sediments vary from 15‰ to 20‰ (Bouquillon et al., 1990), addition of ~6–9% sediment-derived melts can further increase $\delta^{18}\text{O}$ values of the magma by 0.88‰. These proportions of sediment addition have also been shown to account for the Sr-Nd isotope systematics of the Toba volcanic rocks (Liu et al., 2022).

In comparison, $\delta^{11}\text{B}$ values of the rhyolitic melt inclusions (SiO₂ = ~73–79 wt%) hosted in quartz and clinopyroxene of Toba silicic tuffs vary from -7.4‰ to 0.6‰ (Table S3). Previous studies have indicated that the silicic rocks of Toba could have formed by a two-stage AFC of basaltic melts with compositions similar to the Sipisupisu basalts in the lower and upper crust (Liu et al., 2022). It therefore can be inferred that the silicic rocks of Toba could have undergone a similar, but more prolonged, AFC process compared with the basalts. Given that $\delta^{11}\text{B}$ and Nb/B values of the crust underneath Toba are unknown, average crust values are used for our calculation. Results show that AFC alone may not account for the variation of $\delta^{11}\text{B}$ and Nb/B of the rhyolitic melts (Fig. 4). Similar conclusions can be obtained from maintaining constant $\delta^{11}\text{B}$ with increasing SiO₂ contents (Fig. S1). Instead, an increasing amount of AOC and sediment-derived fluids could also have played an important role in the genesis of the Toba silicic melts. Such distinctions between the cogenetic basalts and rhyolites of Toba thus represent either spatial or temporal variations of the large and long-lived volcanic system.

Our study highlights the advantage of combining B and O isotopes to investigate the complex interplay of subduction-zone fluids and melts with the mantle wedge in continental arc settings. Despite extensive fractionation and isotopic compositions of the melt inclusions and early crystallized minerals can record the aggregation of diverse melt batches from both mantle and crust in large trans-crustal arc volcanic systems, a process that is also common in magmatic systems of the Kamchatka arc (eastern Russia) and other arcs worldwide (Iveson et al., 2021).

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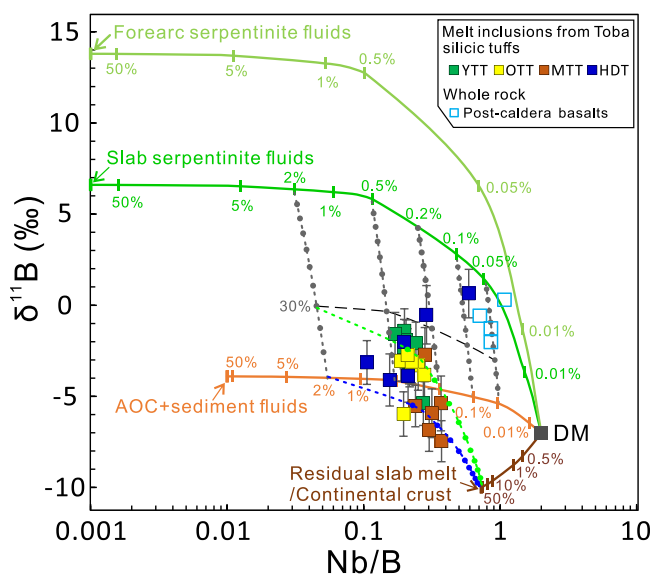


Figure 4. Plot of Nb/B versus $\delta^{11}\text{B}$ for post-caldera basalts and rhyolitic melt inclusions hosted in quartz and clinopyroxene of the Toba volcanic system, Indonesia. Mixing models (solid lines) show contamination of depleted mantle (DM) by fluids derived from forearc serpentinites, slab serpentinites, and AOC + sediments (AOC—altered oceanic crust) at 120 km depth (composition from Tonarini et al., 2011), and by melts from residual slab/continental crust. Gray dot-dash lines indicate composite fluids (0.05–2 wt%) formed by mixing of fluids from slab serpentinites and AOC + sediments.

Blue and green dashed lines show mixing of 2 wt% fluids derived from AOC + sediments with continental crust/slab melts, and 2 wt% fluids derived from slab serpentinites (30%) and AOC + sediments (70%) with continental crust/slab melts, respectively. Each dot on the dashed lines represents a 10% increment. YTT—Youngest Toba tuff; OTT—Oldest Toba tuff; MTT—Middle Toba tuff; HDT—Haranggaol Dacite tuff.

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