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Key Points:

- Fluids derived from serpentinite ([plus minus] altered oceanic crust) have an important contribution to the halogen budget of arc magmas
- Decoupled Sr-Nd isotopes plus heavy boron isotope indicates a combination of recycled serpentinite and altered oceanic crust in arc magmas
- Recycling of serpentinite fluids and fluids from altered oceanic crust are usually coupled in subduction zones

Supporting Information:

- Supporting Information S1
- Data Set S1

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Halogen (F, Cl) Concentrations and Sr-Nd-Pb-B Isotopes of the Basaltic Andesites From the Southern Okinawa Trough: Implications for the Recycling of Subducted Serpentinites

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Abstract Serpentinites are increasingly recognized as playing an important role in the global geochemical cycle. However, discriminating the contributions of serpentinites to arc magmas from those of other subduction components is challenging. The Okinawa Trough is a back-arc basin developed behind the Ryukyu subduction zone, where magmas are extensively affected by sediment subduction. In this study, we reported the F-Cl concentrations and Sr-Nd-Pb-B isotopes of basaltic andesites from the Yaeyama Graben, Yonaguni Graben, and Irabu Knoll in the southern Okinawa Trough. The Irabu Knoll lavas show the most enrichment of fluid-mobile elements and F ± Cl, and have the heaviest B isotopes (δ^{11}B: +6.6 ± 1.5‰). They also have decoupled Sr-Nd isotopes: higher $^{87}Sr/^{86}Sr$ (~0.7049) but have no obvious decrease of $^{143}Nd/^{144}Nd$ (~0.5128). Results from slab dehydration modeling and mixing calculations suggest that the heavy δ^{11}B in the Irabu Knoll lavas is not consistent with fluids derived from altered oceanic crust (AOC), sediments, or wedge serpentinites (formed in the mantle wedge), but rather from slab serpentinites (formed within the subducting plate); sediments control the subduction input of Nd, whereas the decoupled Sr-Nd isotopes are most likely due to the excess radiogenic Sr carried by AOC fluids. Our results imply that recycling of serpentinite fluids and AOC fluids are usually coupled in subduction zones, as the arc lavas influenced by subducted serpentinite generally show Sr-Nd isotopes decoupling. The large variation of Sr-Nd-B isotopes observed in a relatively localized area is consistent with a focused migration through the mantle wedge of components from multiple sources.

1. Introduction

Subduction zone lavas record a significant contribution from lithospheric materials that have been mixed into the mantle at convergent margins. Identifying the mechanism by which this occurs is critical to understanding the mass transfer between crust and mantle as well as the origin of mantle heterogeneity. Altered oceanic crust (AOC) and sediments have long been considered as major subduction components that are identified in arc or back-arc magmas through various elemental and isotopic tracers (e.g., Chauvel et al., 2008; Hauff et al., 2003; Nielsen & Marschall, 2017; Philippot et al., 1998; Plank & Langmuir, 1993; Straub et al., 2004; Turner et al., 2012). Recently, however, the occurrence of serpentinites in subduction zones has drawn much attention and are thought to have an important role in global geochemical cycles (Deschamps et al., 2013). Serpentinites are highly enriched in volatiles (e.g., H_2O, halogens, and noble gases; e.g., Alt et al., 2012; John et al., 2011; Kendrick et al., 2018; Krantz et al., 2019; Pagé et al., 2018; Rüpke et al., 2004) and fluid mobile elements (e.g., B, Li, As, Sb, Pb, U, Cs, Ba; e.g., Deschamps et al., 2010, 2012; Peters et al., 2017; Savov et al., 2005; Scambelluri et al., 2001), which are also enriched in arc lavas (Leeman, 1996; Ryan & Chauvel, 2014). Therefore, serpentinite-derived fluids might have a significant influence on the geochemistry of arc magmas (Hattori & Guillot, 2003; Scambelluri, Fiebig, et al., 2004; Scambelluri et al., 2015; Spandler et al., 2014; Stern et al., 2006; Tonarini et al., 2007).

The lavas in the Ryukyu-Okinawa Trough system are considered to have been significantly influenced by subducted sediments (Shu et al., 2017). Nevertheless, recycled sediments alone cannot account for the compositional variability observed in the lavas of southern Okinawa Trough, that is, a range of incompatible trace elements and Sr isotopic compositions but relatively uniform Nd isotopic ratios (Shinjo et al., 1999). Considering the geochemical features of serpentinite or AOC (e.g., high $^{87}Sr/^{86}Sr$ and high $^{143}Nd/^{144}Nd$; De Hoog & Savov, 2018; Staudigel et al., 1995), the decoupling of Sr-Nd isotopes might be related to the subduction of serpentinite or AOC. The presence of serpentinites in the mantle wedge of the Ryukyu subduction zone has been identified seismically (e.g., high V_P/V_S, strongly anisotropic; Chou et al., 2009; McCormack et al., 2013; Nagaya et al., 2016). However, the presence of sediment- or AOC-derived fluids makes the identification of a "serpentinite signal" in arc magmas difficult because these fluids are also enriched in volatiles and fluid-mobile elements and, thus, are indistinguishable from serpentinite-derived fluids. In addition, subduction-related serpentinites include two types (Martin et al., 2020): (1) slab serpentinites, which are generated by alteration of ultramafic rocks by seawater in the subducting plate; and (2) mantle wedge serpentinites, which form by hydration of the overlying mantle wedge by slab fluids (De Hoog & Savov, 2018; Deschamps et al., 2013; Martin et al., 2016). Discriminating the contributions of the two different types of serpentinites to arc magmas is vital for understanding the roles of serpentinite in geochemical recycling, but is also challenging (Martin et al., 2020).

Boron (B) isotopes ($\delta^{11}B = [(^{11}B/^{10}B)_{Sample}/(^{11}B/^{10}B)_{SRM951}-1]\times1{,}000$) are a powerful tool for tracing subducted serpentinites (e.g., Cannaò et al., 2016; De Hoog & Savov, 2018; Harvey, Garrido, et al., 2014; Martin et al., 2016, 2020; Yamada et al., 2019). First, B is enriched in serpentinites ([B] up to 100 ppm, where brackets indicate concentration) but is extremely depleted in the mantle ([B] < 0.1 ppm) (Leeman, 1996; Marschall, 2018). Second, large differences in $\delta^{11}B$ exist among the slab serpentinites (+5.5‰ to +40.5‰; Martin et al., 2020), mantle wedge serpentinites (−14‰ to +10‰; Martin et al., 2020), sediments (mostly negative; Ishikawa & Nakamura, 1993), AOC (mostly 0 to +5‰; Marschall, 2018), and mantle (−10‰ to −7‰; Marschall, 2018). Thus, B isotopes have been employed to identify the presence of recycled serpentinite in arc magmas (e.g., Bouvier et al., 2019; Cooper et al., 2020; Jones et al., 2014; Leeman et al., 2017; Tonarini et al., 2007, 2011). In this study, we analyzed the Sr-Nd-Pb-B isotopes and halogen (F and Cl) concentrations of basaltic andesites from the Yaeyama Graben, Yonaguni Graben, and Irabu Knoll in the southern Okinawa Trough to (1) test whether B isotopes are still an effective tracer for subducted serpentinite in sediment-dominated settings and (2) constrain the relative importance of subducted serpentinites versus AOC or sediment on the compositional heterogeneity of magmatism in subduction zones.

2. Geological Settings

The Okinawa Trough is a nascent back-arc basin that developed behind the Ryukyu trench-arc system (Figure 1). It is located in the eastern margin of the Asian continent and extends from southwest of Kyushu Island to north of Taiwan Island; it is divided into northern, middle, and southern segments by the Tokara fault and Kerama fault (Figure 1). The back-arc basin started its extension at around middle to late Miocene, as a result of the subduction of the Philippine Sea plate (Lee et al., 1980; Sibuet et al., 1987). The Okinawa Trough is characterized by high heat flow and widespread volcanic activity. Three types of volcanism have been identified in the basin (Sibuet et al., 1998): (1) the modern arc volcanism that is located just east of the Ryukyu Arc; (2) the present-day back-arc volcanism located in the central grabens; and (3) the cross-back-arc volcanism that is manifested as two anomalous volcanic zones in the middle and southernmost part of the trough.

The southern Okinawa Trough lies to the south of the Kerama fault and includes all three types of volcanism mentioned above (Figure 1). The Yaeyama Graben and Yonaguni Graben are two nearly EW trending central grabens that mainly erupt basalts and basaltic andesites (Li et al., 2019; Shinjo et al., 1999; Shu et al., 2017). The Yaeyama Graben is the deepest part of the trough (>2,000 m) and has the thinnest crust (<10 km) and, thus, represents the highest degree of back-arc extension (Arai et al., 2017; Nishizawa et al., 2019). The southern Ryukyu Volcanic Front is adjacent to the Ryukyu Arc and constitutes the southern part of the present-day arc volcanism. The lavas of the volcanic front range from basalts to rhyolites but mostly have SiO_2 concentrations lower than 56 wt.% (Figure 2). The Irabu Knoll, mainly composed of basaltic rocks (Figure 2), is located on the northeast end of the southern Ryukyu Volcanic Front, and the

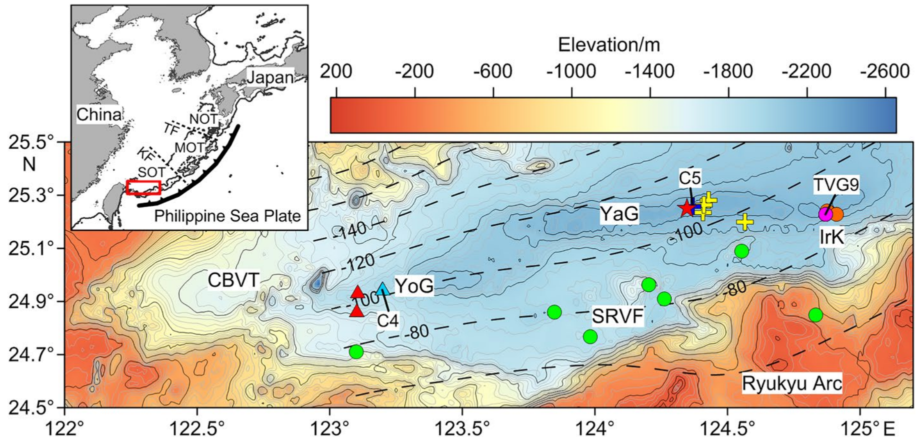

Figure 1. Bathymetric map of the southern Okinawa Trough (SOT) showing the sampling sites and the locations of the Yaeyama Graben (YaG), Yonaguni Graben (YoG), Irabu Knoll (IrK), southern Ryukyu Volcanic Front (SRVF), and Cross Back-arc Volcanic Trail (CBVT). The contour lines represent the depth of Benioff zone (Hayes et al., 2012). The red rectangle in the inset marks the area that the large map covers. The red star marks the location of the Yokosuka hydrothermal site (Miyazaki et al., 2017). KF, Kerama fault; MOT, middle Okinawa Trough; NOT, northern Okinawa Trough; TF, Tokara fault.

eastern edge of the Yaeyama Graben (Fukuba et al., 2015). The Cross Back-arc Volcanic Trail represents an anomalous volcanic zone, consisting of more than 70 submarine volcanoes, on the southwest end of the Okinawa Trough (Sibuet et al., 1998). The volcanic rocks of the Cross Back-arc Volcanic Trail are dominated by dacites and rhyolites and are consistently enriched in Sr-Nd isotopes, which are believed to be the result of crustal contamination (Z. Chen, Zeng, et al., 2019; Shu et al., 2017). Therefore, the Cross Back-arc Volcanic Trail lavas are atypical back-arc volcanic rocks and were not included in this study.

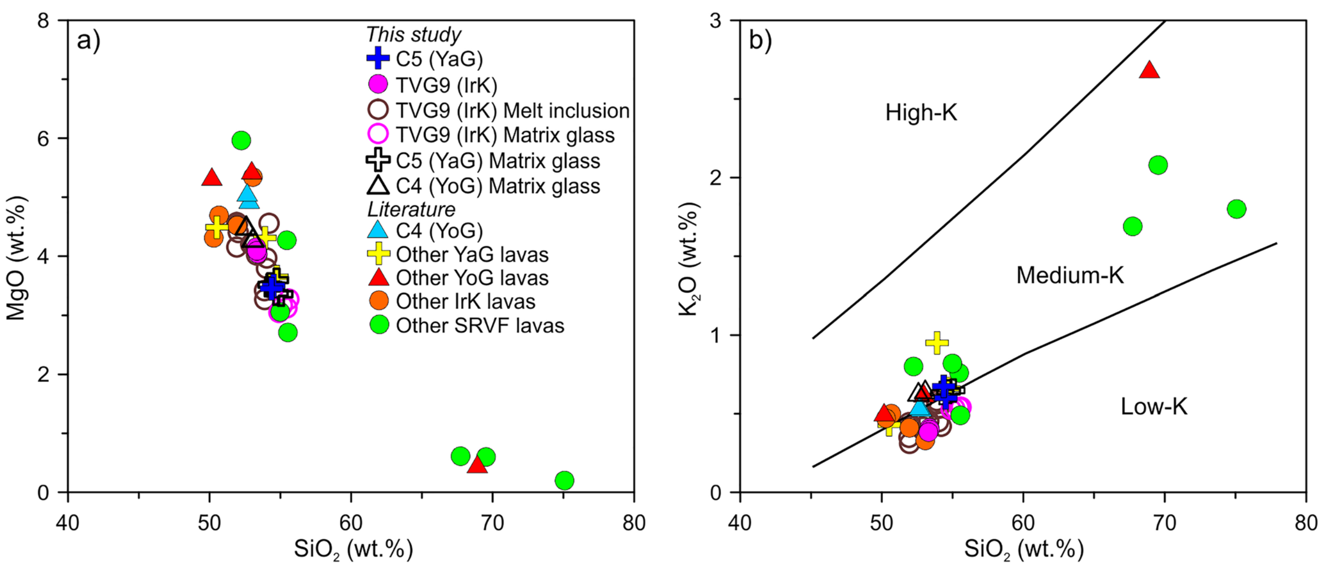

Figure 2. Plots of concentrations of (a) MgO versus SiO_2 and (b) K_2O versus SiO_2 for the volcanic lavas from the southern Okinawa Trough. The major element data of C4 samples are from Z. Chen (2019). Other literature data are from Shu et al. (2017). Boundaries on plot (b) are according to Roberts and Clemens (1993). IrK, Irabu Knoll; SRVF, southern Ryukyu Volcanic Front; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

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3. Samples and Methods

Our samples are fresh basaltic andesites collected from the Yaeyama Graben (C5), Yonaguni Graben (C4), and Irabu Knoll (TVG9), using a television sampler during the "HOBAB4" cruise of R/V KEXUE in 2016. Both the Yaeyama Graben samples (C5) and Yonaguni Graben samples (C4) are vesicular lavas mainly composed of microlites (plagioclase, pyroxene, olivine) and glass (Figures S1a and S1b), implying that the melts ascend rapidly. The Irabu Knoll samples (TVG9) have much higher degree of crystallinity with abundant phenocrysts including plagioclase, orthopyroxene, clinopyroxene, and olivine; some phenocrysts contain glassy melt inclusions (Figures S1c–S1f). Since B and Cl compositions of submarine rocks are easily altered by seawater, whole rock analyses might not be reliable, B isotopic compositions and halogen contents of mineral-hosted melt inclusions and matrix glasses in our samples were measured by secondary ion mass spectrometry (SIMS).

Major elements were determined by X-ray fluorescence spectrometer (XRF, Rigaku RIX 2000) on lithium tetraborate fused glass discs at the analytical laboratory of Beijing Research Institute of Uranium Geology (BRIUG). Trace elements were determined using an inductively coupled plasma mass spectrometry (ICP-MS, ELAB DRC II) at the Institute of Oceanology, Chinese Academy of Sciences (IOCAS). Sr, Nd and Pb isotopic ratios were determined using thermal ionization mass spectrometry (TIMS; MAT-262) at the Radiogenic Isotope Laboratory of the University of Science and Technology of China (USTC). Boron isotopic compositions and halogen (F, Cl) concentrations of the matrix glasses and melt inclusions were determined by SIMS using the Cameca IMS 1280 ion microprobe at the Northeast National Ion Microprobe Facility (NENIMF) at the Woods Hole Oceanographic Institution (WHOI). Major elements of the matrix glasses and melt inclusions were determined by electron microprobe (EMPA, JEOL JXA-8230) at the Ocean University of China. The detailed sample preparation and analytical procedures are given in Supporting Information.

4. Results

The major elements, trace elements, and Sr-Nd-Pb isotopic compositions for lavas from the Yaeyama Graben, Yonaguni Graben, Irabu Knoll, and southern Ryukyu Volcanic Front are given in Tables S1–S3. The data of all C4 samples and Sr-Nd-Pb isotopic data of C5 and TVG 9 samples have been reported by Z. Chen (2019). This data set also includes the previous data of the research areas (Shu et al., 2017). Most of the lavas from the southern Okinawa Trough have low MgO concentrations (<6 wt.%) and medium to low K_2O concentrations (Figure 2). C5, C4, and TVG9 samples have similar major element compositions (Figure 2). The major element compositions of melt inclusions in TVG9 samples resemble those of whole rock; the matrix glasses in TVG9 samples are slightly more evolved, whereas those in C5 and C4 samples are parallel to whole rock (Table S1; Figure 2).

C5, C4, and TVG9 samples have distinct rare earth element (REE) and trace element patterns (Figure 3, where only data of basalts and basaltic andesites are presented). C5 and other Yaeyama Graben samples are characterized by high concentrations of incompatible trace elements and slightly light REE (LREE) enriched patterns. TVG9 and other Irabu Knoll samples are also slightly enriched in the LREE distributions, but they have systematically lower trace element concentrations, with the exception of Pb. C4 samples and other Yonaguni Graben samples have intermediate trace element concentrations and REE that are more fractionated: the concentrations of light REE (e.g., La, Ce) are comparable to those of Yaeyama Graben lavas, while the concentrations of heavy REE (HREE; e.g., Er, Tm, Yb, Lu) are similar to those of Irabu Knoll lavas.

The Sr-Nd-Pb isotopic compositions of the lavas become increasingly enriched from the Yaeyama Graben to the Yonaguni Graben, and to the southern Ryukyu Volcanic Front (Figure 4). C5 samples have the least radiogenic Sr and Nd ($^{87}Sr/^{86}Sr$ = 0.703714–0.703726, $^{143}Nd/^{144}Nd$ = 0.512946–0.512956) among the southern Okinawa Trough lavas. TVG9 samples and some other Irabu Knoll lavas have higher $^{87}Sr/^{86}Sr$ (>0.7046), but they also have relatively high $^{143}Nd/^{144}Nd$ ratios (~0.5128). On Sr-Nd isotope plots, the data of Yaeyama Graben and Yonaguni Graben lavas plot on a mixing line between the mantle and sediments, whereas the data of Irabu Knoll lavas mostly plot above the mixing line (Figure 4a). Compared to the volcanic rocks from the middle Okinawa Trough (MOT), the southern Okinawa Trough lavas have more enriched Pb isotopic compositions (Figures 4b and 4c), showing a DUPAL-like signature (Y. Zhang et al., 2018). On the

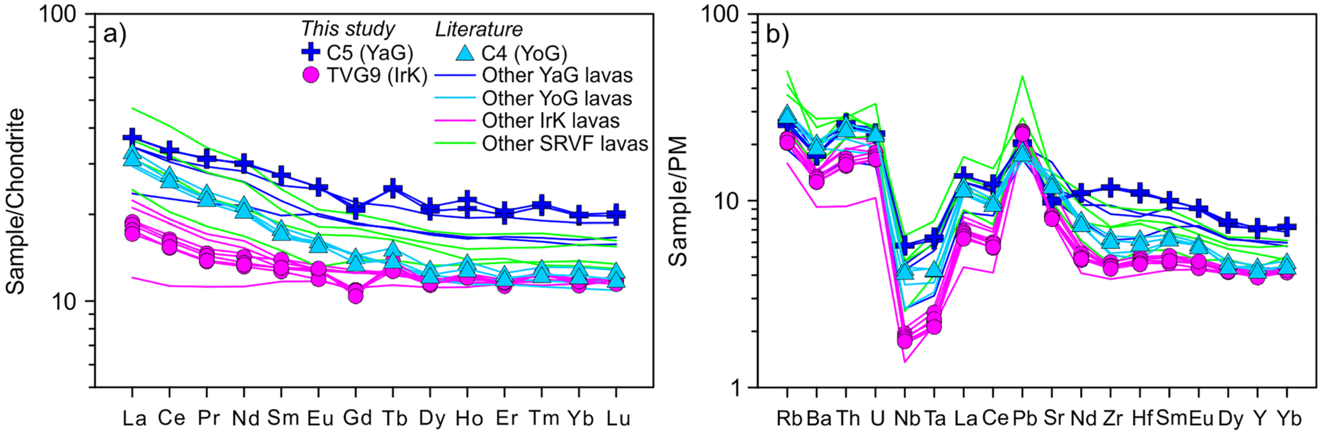

Figure 3. (a) Chondrite normalized rare earth element (REE) patterns and (b) primitive mantle (PM) normalized trace element patterns for the southern Okinawa Trough lavas (SiO_2 < 56 wt.%). The trace element data of C4 samples are from Z. Chen (2019). Other literature data are from Shu et al. (2017). Primitive mantle and chondrite data are from McDonough and Sun (1995). IrK, Irabu Knoll; SRVF, southern Ryukyu Volcanic Front; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

Pb-Pb isotopic plots, the southern Okinawa Trough lavas define a linear trend toward detrital sediments (Figures 4b and 4c).

The B and halogen data are reported in Tables S4 and S5 and are shown in Figures 5 and 6. The southern Okinawa Trough lavas have a large range in $\delta^{11}B$ values, increasing from −4.3‰ to −2.8‰ in Yaeyama Graben lavas (C5) to +0.9‰ to +1.7‰ in Yonaguni Graben lavas (C4), and to +5‰ to +9.1‰ for Irabu Knoll lavas (TVG9). The approximated B concentrations of these samples increase with increasing $\delta^{11}B$ (Figure S4). Note that the B isotopic compositions of the melt inclusion and the matrix glasses are the same for TVG9 (Figure 5). The Cl and F concentrations of the matrix glasses ([Cl] = 740–884 ppm, [F] = 232–297 ppm) are also indistinguishable with those of melt inclusions ([Cl] = 711–940 ppm, [F] = 215–335 ppm) for TVG9 (Table S5). The C5 matrix glasses have much higher Cl (1,520–1,748 ppm) and F (370–413 ppm) concentrations than the C4 and TVG9 glasses. The C4 matrix glasses have slightly lower Cl concentrations (592–698 ppm) and higher F concentrations (282–334 ppm) than TVG9 glasses.

It is noted that all the lavas in this study are evolved relative to primary partial melts of the mantle. Magma differentiation has little effect on Sr-Nd-Pb-B isotopes, but can significantly influence the concentrations of trace elements and volatiles. In the following discussions, we (1) selected the data with SiO_2 < 56 wt.% and (2) utilized element ratios instead of concentrations to minimize this effect. In such case, the B and halogen concentrations in the melt inclusions were not corrected for post-entrapment crystallization (PEC), which has little effect on elemental ratios (e.g., Cl/K) and isotopic compositions (e.g., $\delta^{11}B$).

5. Discussion

5.1. Subduction Input of Halogens to the Mantle of Southern Okinawa Trough

Lavas recovered from different parts of the southern Okinawa Trough define elemental and isotopic variability that might be attributed to either crustal contamination or addition of a subduction component to their source region. Results from previous studies indicate that southern Okinawa Trough lavas (with the exception of those from the Cross Back-arc Volcanic Trail) are free from the influence of crustal contamination (e.g., Shinjo et al., 1999; Shu et al., 2017). The lack of any correlation between the MgO concentration and either Sr or Nd isotopes in the southern Okinawa Trough lavas also indicates a lack of crustal contamination (Figure S5). The Sr, Nd, and Pb isotopes show overall enrichment approaching the trench (Figure 4), indicating that regional geochemical trends recorded in the southern Okinawa Trough lavas are most likely controlled by contribution of subduction components. Fluids derived from subducted sediments, AOC, and serpentinized lithospheric mantle control the volatile flux in subduction zones (Jarrard, 2003; Philippot et al., 1998; Straub & Layne, 2003; Wallace, 2005). In order to trace the influence of subduction fluids, we used the ratios of fluid mobile (e.g., Rb, Ba, Pb, Cs) to fluid immobile trace elements (e.g., Th, La, Ce), which

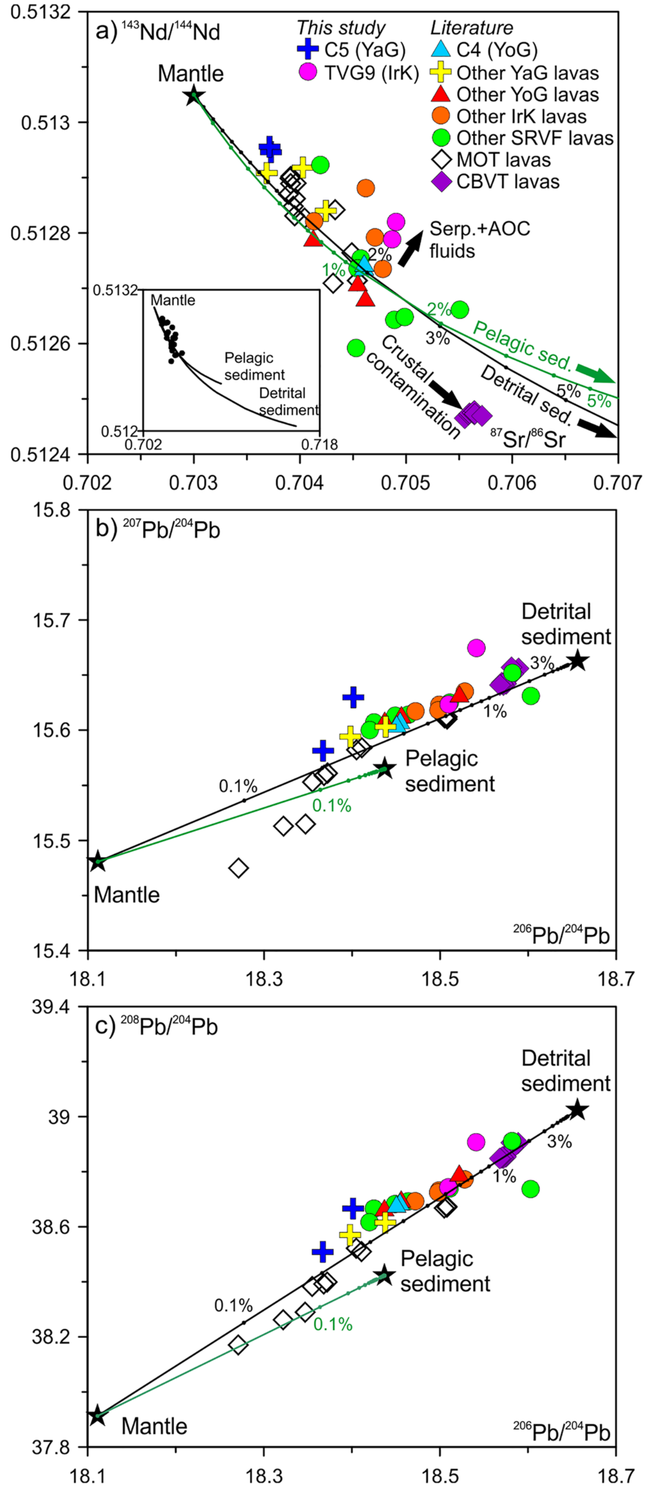

Figure 4. Plots of (a) $^{87}Sr/^{86}Sr$ versus $^{143}Nd/^{144}Nd$, (b) $^{207}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$, and (c) $^{208}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ for the southern Okinawa Trough lavas. The data of middle Okinawa Trough (MOT) lavas (mainly basaltic rocks) are from Hoang and Uto (2006) and Shu et al. (2017). The data of mantle are from Nielsen and Marschall (2017) and Y. Zhang et al. (2018). The data of Cross Back-arc Volcanic Trail (CBVT) lavas and pelagic sediment outboard of the middle Ryukyu Arc are from Shu et al. (2017). The data of detrital sediment outboard of the southern Ryukyu Arc are from the weighted average compositions of a sediment core (VM28-313) in the southernmost part of the Ryukyu trench (Bentahila et al., 2008). Other data sources are shown in Figure 2 and Table S3. The inset in (a) shows the full mixing lines between mantle with detrital sediment and pelagic sediment. IrK, Irabu Knoll; SRVF, southern Ryukyu Volcanic Front; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

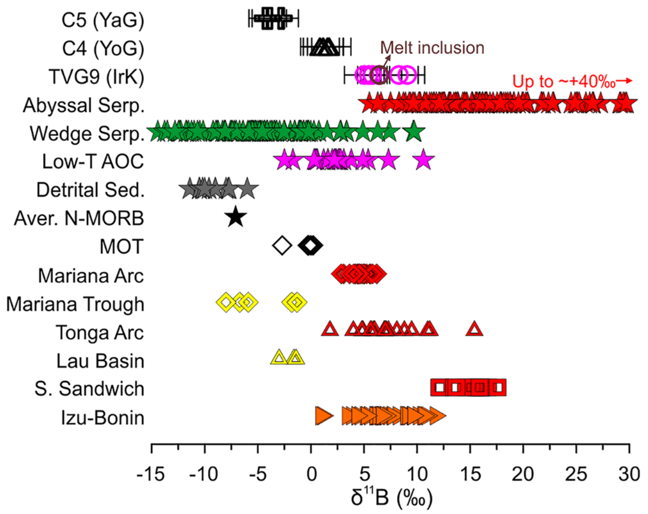

Figure 5. Boron isotope ($\delta^{11}B$) compositions of the matrix glasses and melt inclusion in the southern Okinawa Trough lavas. The error bars indicate 1σ uncertainties. Also shown are the data of some arcs, back-arcs, serpentinite, AOC, MORB, and sediment for comparison. Data sources: Abyssal serpentinite (Boschi et al., 2008, 2013; Harvey, Savov, et al., 2014; Martin et al., 2016, 2020; Vils et al., 2009), Mantle wedge serpentinite (Martin et al., 2016, 2020), Low-T AOC (Ishikawa & Nakamura, 1992; Smith et al., 1995), Detrital sediment (Chetelat et al., 2009), Average N-MORB (Marschall, 2018), middle Okinawa Trough (MOT) (Pi et al., 2016), Mariana Arc (Ishikawa & Tera, 1999), Mariana Trough and Lau Basin (Chaussidon & Jambon, 1994; Chaussidon & Marty, 1995), South Sandwich (Tonarini et al., 2011), Izu-Bonin (Ishikawa & Nakamura, 1994; Straub & Layne, 2002, 2003). IrK, Irabu Knoll; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

are not significantly fractionated by either partial melting or magma differentiation (Ryan & Chauvel, 2014; Zamboni et al., 2016). The Yaeyama Graben lavas have the highest trace element concentrations (Figure 3) but the lowest fluid mobile/immobile trace element ratios (Figure 6), indicating minimal input of subduction fluids to their source region. This is consistent with their location farthest from the trench (Figure 1), and also consistent with their low $\delta^{11}B$ values (Figure 5) and depleted Sr, Nd, and Pb isotopes (Figure 4). The fluid mobile/immobile trace element ratios increase progressively from the Yaeyama Graben to the Yonaguni Graben lavas, and to the southern Ryukyu Volcanic Front lavas (Figure 6), with the highest ratios recorded by the Irabu Knoll lavas, indicating that the Irabu Knoll lavas might have had the most subduction fluids incorporated into their source region.

F and Cl are important volatile components in subduction-related magmas (e.g., Churikova et al., 2007; Fischer, 2008; Rose-Koga et al., 2014; Straub & Layne, 2003; Villemant & Boudon, 1999). Subducted sediments ([F] up to ~1,250 ppm, [Cl] up to ~2,000 ppm), AOC ([F] = ~207 ppm, [Cl] = ~216 ppm), and serpentinites ([F] = ~204 ppm, [Cl] = ~2,000 ppm) have much higher F and Cl concentrations (Barnes et al., 2018 and references therein) than mantle peridotite ([F] = ~12 ppm, [Cl] = ~5 ppm) (Kendrick et al., 2018). In addition, F and Cl are less influenced by magma degassing as they have higher solubilities in the melt than CO_2 and H_2O at lower pressure (Webster, 1997; Webster et al., 1999). Therefore, the high F and Cl abundances in subduction zone magmas indicate a contribution of volatile-rich, slab-derived agents to the mantle (Bénard et al., 2017; Bouvier et al., 2008, 2010, 2019; Kendrick et al., 2011, 2012; Pagé & Hattori, 2019; Portnyagin et al., 2007; Rose-Koga et al., 2012, 2014; Straub & Layne, 2003).

Cl has a similar incompatibility to K (Kent et al., 2002) and F has a similar incompatibility as Nd (Y. Chen et al., 2015). In order to minimize the influence of partial melting or crystallization differentiation, we used Cl/K and F/Nd to assess the halogen

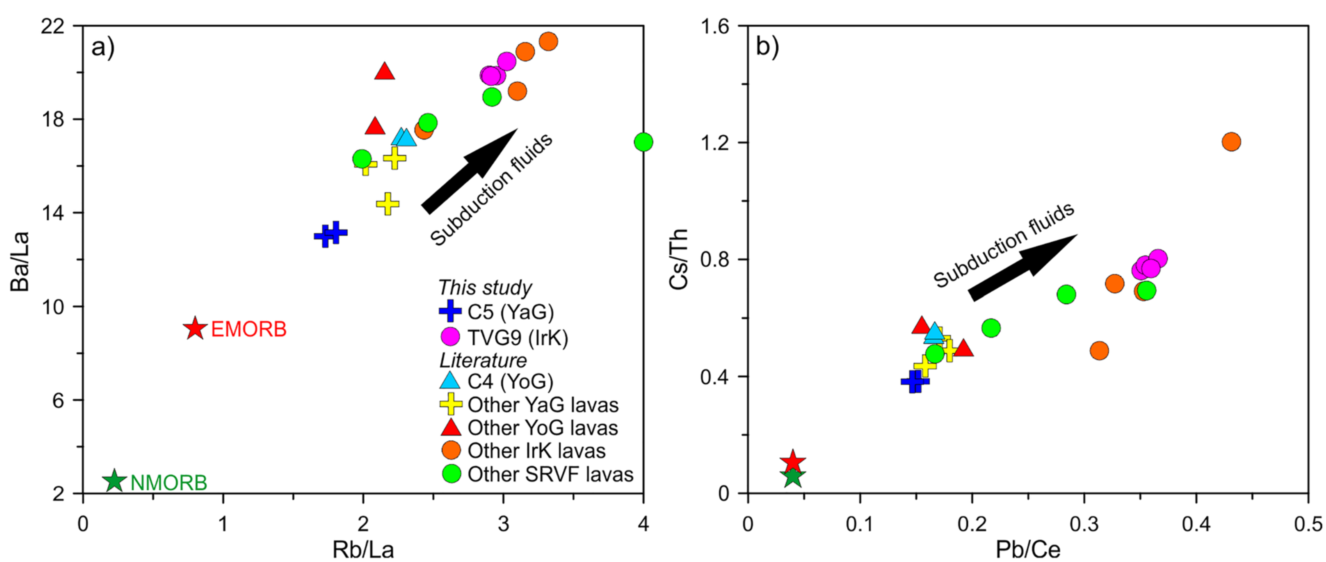

Figure 6. Plots of (a) Ba/La versus Rb/La and (b) Cs/Th versus Pb/Ce for the southern Okinawa Trough lavas. The black arrows point to the increasing contributions of subducted fluids. The data of NMORB and EMORB are from Sun and McDonough (1989). IrK, Irabu Knoll; SRVF, southern Ryukyu Volcanic Front; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

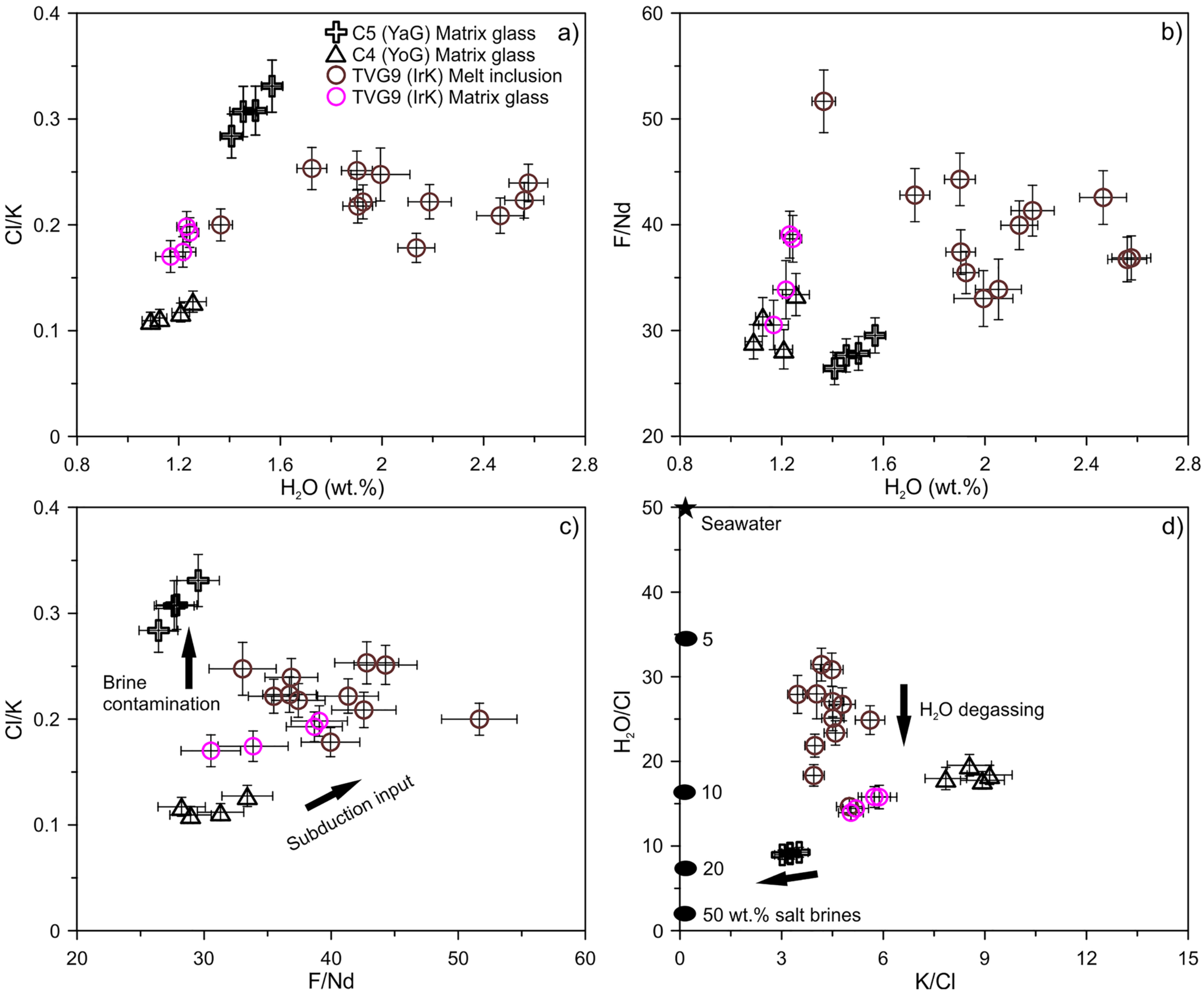

Figure 7. Plots of (a) Cl/K versus H_2O, (b) F/Nd versus H_2O, (c) Cl/K versus F/Nd, and (d) H_2O/Cl versus K/Cl. The error bars indicate 2σ uncertainties. The Yaeyama Graben (C5) lavas have much higher Cl/K ratio but lower F/Nd ratio, suggesting the brine contamination. The data of seawater and brines are from Kendrick et al. (2015). IrK, Irabu Knoll; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

concentrations in the magma source of the southern Okinawa Trough (Figure 7). The Cl/K and F/Nd ratios do not vary with decreasing water concentration among the melt inclusions and matrix glasses of TVG9 samples (Figures 7a and 7b), implying that the Cl and F concentrations in the melt were not influenced by magma degassing. The Yaeyama Graben lavas show higher Cl/K ratios than the Yonaguni Graben and Irabu Knoll lavas, which is obviously not a result of more subduction input for the Yaeyama Graben lavas, as they have the lowest F/Nd (Figure 7) and lowest ratios of fluid mobile elements to fluid immobile elements (Figure 6), and have the least radiogenic Sr, Nd, and Pb (Figure 4). This indicates that other Cl-rich components might have been incorporated into these lavas. One possibility is that the Yaeyama Graben lavas were influenced by seawater alteration. However, the Yaeyama Graben lavas are fresh (LOI = 0.3–0.6 wt.%). In addition, they have very low $\delta^{11}B$ values and $^{87}Sr/^{86}Sr$ ratios (Figures 4a and 5), suggesting minor seawater alteration. Assimilation of pure seawater ($H_2O/Cl \approx 50$) by magma is also unlikely, as it cannot explain the lower H_2O/Cl ratios (≈9) of the Yaeyama Graben lavas compared to the Yonaguni Graben and Irabu Knoll lavas (Figure 7d) (Kendrick et al., 2013, 2015; Kent, Norman, et al., 1999). Thus, a more plausible explanation is that the melt was contaminated by highly saline brines prior to or during eruption (Broadley et al., 2017; Coombs et al., 2004; Kendrick et al., 2015; Kent, Clague, et al., 1999; Kent et al., 2002;

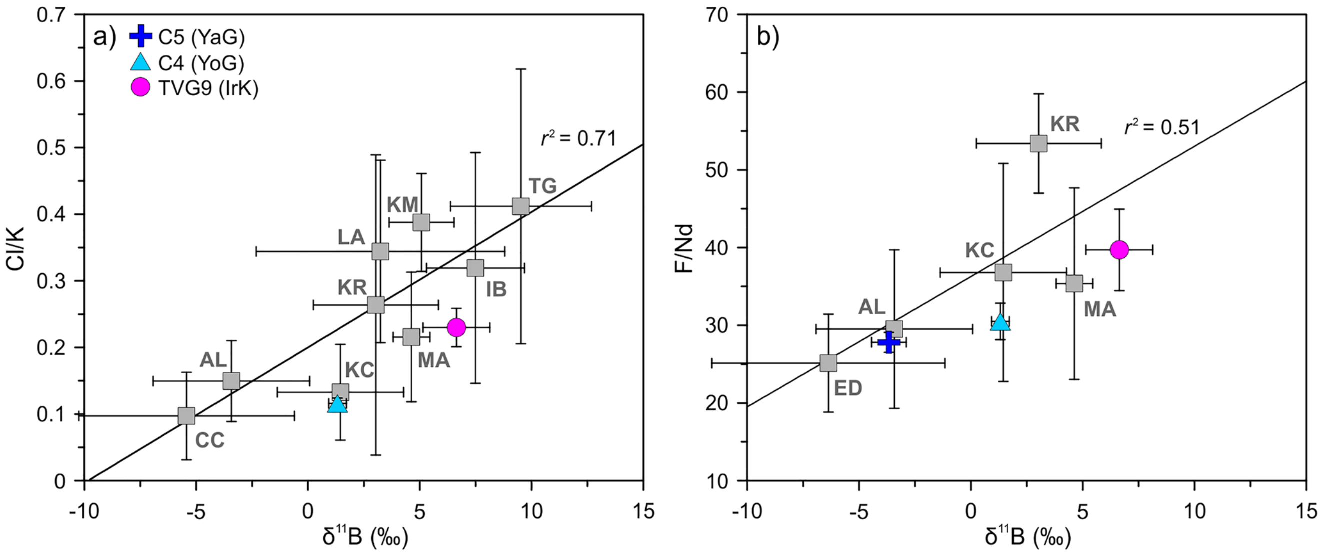

Figure 8. Plots of (a) Cl/K versus δ^{11}B and (b) F/Nd versus δ^{11}B for the lavas from southern Okinawa Trough lavas and 10 modern arcs (from GEOROC database: http://georoc.mpch-mainz.gwdg.de/georoc/ and Cooper et al., 2020; data with MgO > 4 wt.% were adopted). Each gray square represents the average values of each individual arc. The error bars indicate 1σ uncertainties. The arc average values are given Table S6 and the source data are given in Datasets. The linear positive correlations between δ^{11}B and Cl/K, F/Nd for global arcs suggest serpentinite- (±AOC-) derived fluids probably dominate the subduction input of halogens into the sub-arc mantle. AL, Aeolian; CC, Cascades; ED, Ecuador; IB, Izu-Bonin; IrK, Irabu Knoll; KC, Kamchatka; KM, Kermadec; KR, Kurile; LA, Lesser Antilles; MA, Mariana; TG, Tonga; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

Kent, Norman, et al., 1999). Cl-rich brine can be generated via the phase separation of hydrothermal fluids (Coumou et al., 2009). A new high temperature hydrothermal site (Yokosuka) has been discovered in the Yaeyama Graben (Miyazaki et al., 2017), near the sampling site of C5 (Figure 1). The hydrothermal fluid is characterized by low Cl concentrations, which is due to subcritical phase separation (Miyazaki et al., 2017), making it possible that highly saline brine exists in the crust of the Yaeyama Graben. If we exclude the possible brine assimilation for the Yaeyama Graben lavas, the Irabu Knoll lavas show the highest enrichment of F ± Cl (Figure 7c), consistent with the higher ratios of fluid mobile elements to fluid immobile elements (Figure 6). Our results are also consistent with a global trend that Cl/K and F/Nd of arc lavas correlate positively with δ^{11}B (Figure 8). Thus, serpentinite- (±AOC-) derived fluids probably represent an important contribution to the halogen budget in the southern Okinawa Trough.

5.2. Identifying the Recycled Serpentinites in the Southern Okinawa Trough Lavas

The Irabu Knoll lavas show the greatest enrichment of fluid mobile elements and halogens, and heaviest B isotopes (δ^{11}B = +6.6 ± 1.5‰) in the southern Okinawa Trough, suggesting that they might have been influenced by serpentinite-derived fluids. However, before drawing this conclusion, other possibilities that lead to heavy B isotopes, such as seawater alteration or the addition of AOC- or sediment-derived fluids, should be excluded.

Seawater has extremely heavy B isotopes (δ^{11}B ≈ +39.6‰; Spivack & Edmond, 1987). In theory, seawater alteration can increase the B isotopic ratios of the altered rocks. However, the Irabu Knoll samples are quite fresh (LOI < 0.5 wt.%). In addition, we measured the δ^{11}B of a clinopyroxene-hosted melt inclusion in the Irabu Knoll lavas, and it is indistinguishable from those of the matrix glasses (Figure 5). We also measured in situ Sr isotopic compositions of plagioclases in the Irabu Knoll lavas, showing homogeneous values from the core to the rim (Figure S6), similar to the whole-rock Sr isotopic ratios. Furthermore, the Cl/K ratios of the matrix glasses are parallel to those of the melt inclusions in the Irabu Knoll lavas (Figures 6a and 6b), suggesting little or no seawater alteration. Thus the relatively heavy B isotopes in Irabu Knoll lavas are unlikely to be due to seawater alteration. In addition, assimilation of high-temperature altered crustal rocks of the Okinawa Trough during magma ascent is also unlikely, as high-temperature altered crustal rocks

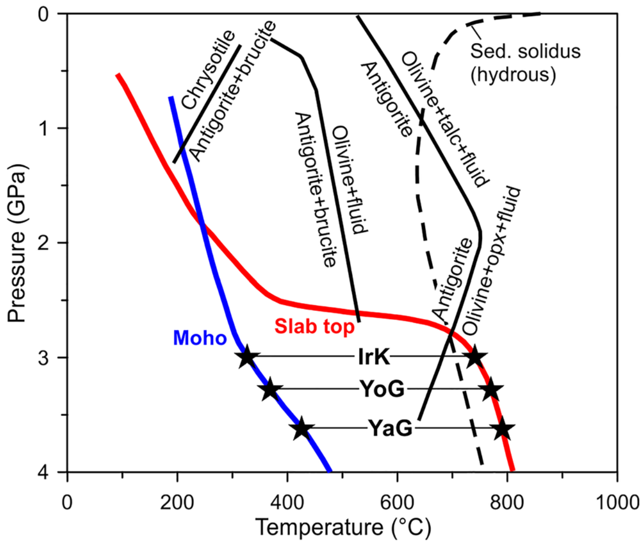

Figure 9. P-T files of the slab top (red line) and Moho (blue line) in the Ryukyu subduction zone, which are according to the "D80" model of Syracuse et al. (2010). The phase boundaries are from Scambelluri, Fiebig, et al. (2004) and Hermann and Spandler (2008). The black stars indicate the P-T conditions of the slab surface and Moho beneath the Irabu Knoll (IrK), Yonaguni Graben (YoG), and Yaeyama Graben (YaG), respectively, based on the slab depth model of Hayes et al. (2012).

would have very low B concentrations (<10 ppm), although they can record heavy B isotopes (Marschall, 2018) and thus it cannot account for the high B concentrations of Irabu Knoll lavas (~40 ppm).

Most of the AOC outboard of the Ryukyu Arc have low Tl isotopes ($\varepsilon^{205}Tl_{aver.} = -5.5‰$) (Shu et al., 2017), corresponding to low temperature AOC (Coggon et al., 2014), which is characterized by relatively light B isotopes ($\delta^{11}B = 0$ to $+5‰$; Marschall, 2018). The sediments entering the sub-arc mantle in the southern Ryukyu area are mainly terrigenous detrital sediments (Shu et al., 2017), as confirmed by the observations that all the southern Okinawa Trough lavas plot on the mantle-detrital sediment mixing lines on the Pb-Pb isotopic plots (Figures 4b and 4c). Detrital sediments are characterized by very low $\delta^{11}B$ values ($-13‰$ to $-8‰$; Chetelat et al., 2009; Ishikawa & Nakamura, 1993). Furthermore, B isotopic fractionation would occur with the progressive slab dehydration as ^{11}B is preferentially partitioned into aqueous fluids (Hervig et al., 2002; King et al., 2007; Pabst et al., 2012; You et al., 1995). Thus, the fluids released from the slab (AOC + sediment) at Benioff zone depth (~90 km) beneath the Irabu Knoll are predicted to have even lower $\delta^{11}B$ values ($\sim+3.1‰$ for AOC fluids and $\sim-8.5‰$ for sediment fluids) according to a slab dehydration model (AOC:Sediments = 9:1) (Figures S7). The model follows the methods of Tonarini et al. (2011) and Jones et al. (2014), in which the release of B from the slab is coupled with water loss (Marschall et al., 2006). The details about the modeling are given in the supporting information (Figure S7). Therefore, the AOC- and sediment-derived fluids cannot account for the heavy B isotopes observed in the Irabu Knoll lavas.

The serpentinites formed in the mantle wedge are a potential fluid source for arc magmas (Martin et al., 2016, 2020). However, recent studies have shown that the mantle wedge serpentinites have relatively light B isotopes ($\delta^{11}B = -14.5‰$ to $+10‰$), as the slab-derived metamorphic fluids have decreasing $\delta^{11}B$ values with increasing depth (Martin et al., 2016, 2020; Yamada et al., 2019). Furthermore, since the subducting Philippine Sea Plate is relatively young and warm, the serpentinites near the slab surface would totally breakdown before reaching the depth below the southern Okinawa Trough region (red line in Figure 9; Syracuse et al., 2010). In comparison, the slab serpentinites have heavy B isotopes ($\delta^{11}B$ up to $+40.5‰$) (e.g., Boschi et al., 2008; Cannaò et al., 2016; Harvey, Savov, et al., 2014; Martin et al., 2020; Vils et al., 2009). Serpentinization can occur beneath the crust formed at intermediate and fast spreading centers in the Pacific as a result of seawater penetration through the slab bending faults outboard of subduction zones (Alt et al., 2012). Thus, such serpentinites can be insulated within the subducting slab and dehydrate at greater depth (Rüpke et al., 2004). The serpentine minerals within the slab are at P-T conditions of the chrysotile-antigorite transition beneath the Irabu Knoll (blue line in Figure 9), and chrysotile breakdown can release significant amounts of ^{11}B-rich fluid (Harvey, Garrido, et al., 2014; Scambelluri, Muntener, et al., 2004).

Above all, the heavy B isotopes of the Irabu Knoll lavas most likely reflect the contribution of slab serpentinites (Figure 10). Some intra-oceanic arcs are characterized by heavy B isotopes, for example, the Mariana Arc ($\delta^{11}B = +2.9‰$ to $+6.2‰$; Ishikawa & Tera, 1999), the South Sandwich Arc ($\delta^{11}B = +12.1‰$ to $+17.6‰$; Tonarini et al., 2011), and the Tonga Arc ($\delta^{11}B = +5.7‰$ to $+15.4‰$; Leeman et al., 2017) (Figure 5). Such positive $\delta^{11}B$ values can only be explained by the contribution of ^{11}B-rich serpentinite fluids released from the slab or serpentinites eroded from serpentinized forearc mantle (cold nose) (Benton et al., 2001; De Hoog & Savov, 2018; Leeman et al., 2017; Ryan & Chauvel, 2014; Tonarini et al., 2011). These arc lavas are influenced by relatively less sediment input (e.g., Leeman et al., 2017; Straub et al., 2015; Tonarini et al., 2011). Our results indicate that B isotopes are still good tracers for the presence of fluids released by dehydration of subducted serpentinites in sediment-dominant settings.

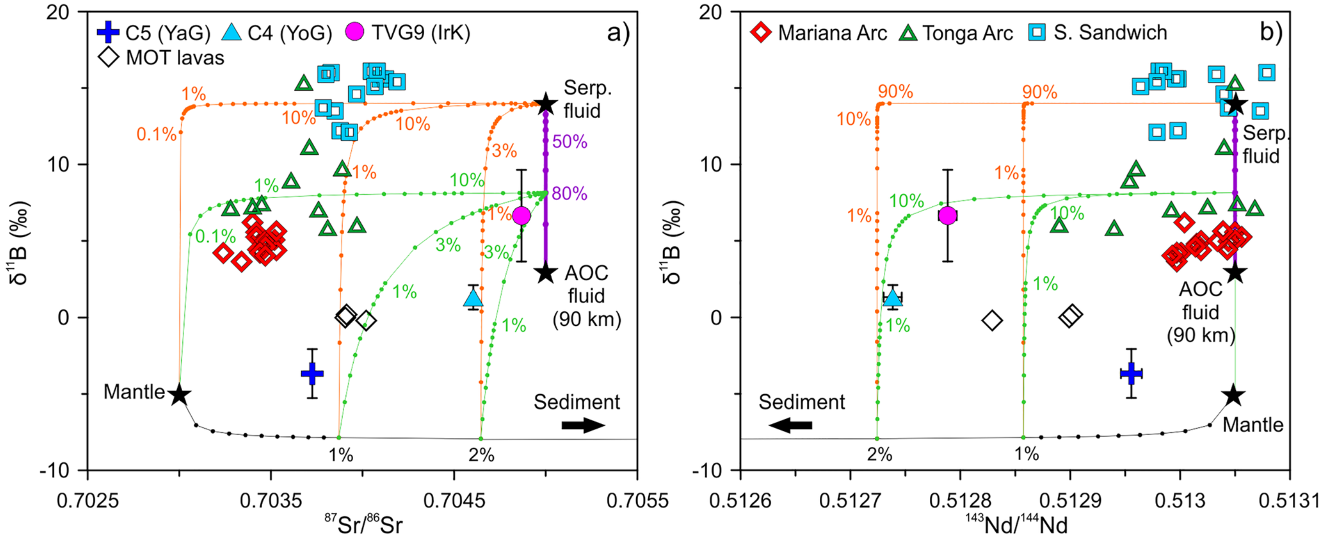

Figure 10. Plot of (a) $\delta^{11}B$ versus $^{87}Sr/^{86}Sr$ and (b) $\delta^{11}B$ versus $^{143}Nd/^{144}Nd$ for the southern Okinawa Trough lavas. Data of the Mariana Arc, Tonga Arc, and South Sandwich Arc are presented for comparison; their data sources are shown in Figure 5. The boron isotope data are average values of multiple analyses on the matrix glasses and melt inclusion (Table S4). The error bars indicate 2σ uncertainties. Error bars on the Sr isotopes are smaller than the symbol size. The mixing lines describe the compositions of mantle source metasomatized by variable proportions of bulk sediment and fluids derived from serpentinite and AOC. The endmember compositions are given in Table 1. IrK, Irabu Knoll; MOT, middle Okinawa Trough; YaG, Yaeyama Graben; YoG, Yonaguni Graben.

5.3. Origin of Sr-Nd Isotopes Decoupling

Subducted sediments play a significant role in the formation of arc magma (e.g., Plank & Langmuir, 1993, 1998). The global arc lava data generally plot on a mixing line between the mantle and the sediments on the Sr-Nd isotopic plots, forming a trend similar to the "mantle array" (Ben Othman et al., 1989; Nielsen & Marschall, 2017; White & Patchett, 1984). As illustrated in Figure 4a, the Yaeyama Graben and Yonaguni Graben lavas plot on the mantle-sediment mixing line, whereas some Irabu Knoll lavas plot on the right side of the mixing line, showing a relative Sr-Nd isotopes decoupling trend (Figure 4a), which is a well-known feature common in many arcs, like the Central America Arc (Tonarini et al., 2007) and South Sandwich Arc (Tonarini et al., 2011; Figure 11). Shinjo et al. (1999) reported this phenomenon in the southern Okinawa Trough and they attributed it to a heterogeneous mantle source. Nevertheless, they fail to give further explanations for the origin of the decoupled Sr-Nd isotopes observed in the Irabu Knoll lavas. The Irabu Knoll lavas show the greatest enrichment of Sr-Nd-Pb isotopes (Figure 4), which is consistent with the incorporation of a significant amount of detrital sediments (1%–2%) as bulk (or as melt, see below) into their source region (Shu et al., 2017). However, sediments alone cannot account for the Sr-Nd isotope decoupling, suggesting that the Irabu Knoll lavas must have been influenced by other processes, for example, (1) alteration of the volcanic rocks by seawater and (2) addition of subduction components with unusually radiogenic Sr into the magma sources, such as fluids derived from sediments, AOC, or serpentinites.

Seawater altered rocks can obtain high $^{87}Sr/^{86}Sr$ from seawater while retaining the original Nd isotopes of the protolith (e.g., Staudigel et al., 1995). However, our samples are free from seawater alteration (see Section 5.2). As previously discussed, the Irabu Knoll lavas have been influenced by subducted slab serpentinite fluids, which are characterized by high $^{87}Sr/^{86}Sr$ (up to ~0.709) (De Hoog & Savov, 2018), and thus are seemingly good candidates for causing Sr-Nd isotope decoupling. However, serpentinite fluids alone can increase $\delta^{11}B$ dramatically but are unable to significantly increase $^{87}Sr/^{86}Sr$ for their relatively low concentrations of Sr (~40 ppm) (Figure 10) (Tonarini et al., 2007). Contributions from other subduction components are, therefore, required.

Fluids from dehydrating sediments have much higher Sr/Nd than the bulk sediments. Addition of such sediment fluids to the mantle source would produce a mixing curve with different curvature than that of the mixing line between mantle and bulk sediments (Nielsen & Marschall, 2017). Nevertheless, based on the thermal structure of the Ryukyu subduction zone, the sediments are expected to be melted beneath

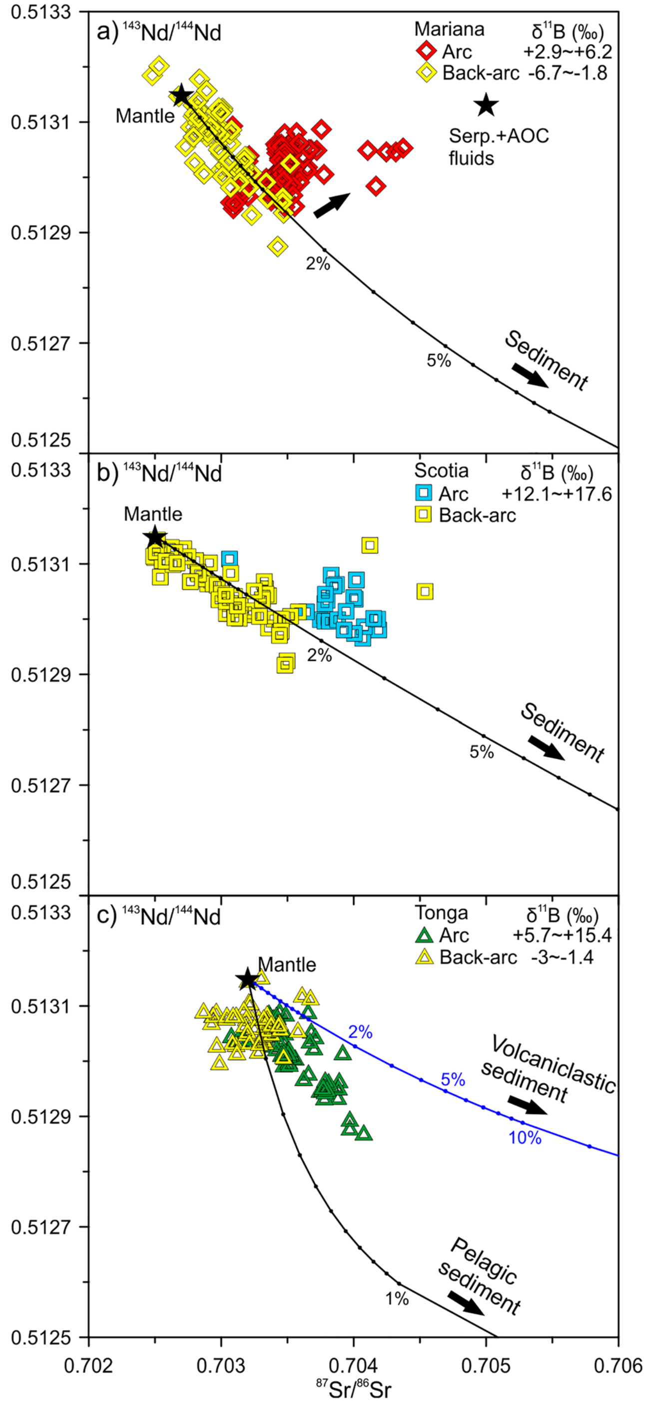

Figure 11. Plots of $^{87}Sr/^{86}Sr$ versus $^{143}Nd/^{144}Nd$ for arc and back-arc lavas in the (a) Mariana, (b) Scotia, and (c) Tonga subduction zones. The plot data are from GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/). The mixing endmembers (mantle and sediments) are according to Nielsen and Marschall (2017). The data of serpentinite and AOC are from De Hoog and Savov (2018) and Staudigel et al. (1995), respectively. The source of B isotope data are shown in Figure 5. Considering the complex geology of the Lau Basin, the data from the Central Lau Spreading Center, Eastern Lau Spreading Center, and Valu Fa Ridge are used as the back-arc data. Although the sediments outboard of the Tonga Arc include two types (Nielsen & Marschall, 2017), the back-arc lavas mostly plot near the mixing line defined between mantle and pelagic sediment, suggesting that pelagic sediment is a major subduction input. In such case, the Tonga Arc data show Sr-Nd isotopes decoupling feature.

Table 1
Parameters for Mixing Models

	$\delta^{11}B$ (‰)	B (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{143}Nd/^{144}Nd$	Nd (ppm)
Mantle[a]	−5	0.03	0.703	9.7	0.51305	0.7
Serpentinite fluid[a]	14	270	0.705	40	0.51305	0.004
AOC fluid (90 km)[b]	3	77	0.705	358	0.51305	1.1
Detrital sediment[c]	−8	65	0.715842	69.9	0.51204	16.33

[a]From Tonarini et al. (2007). [b]Calculated from the slab dehydration model (AOC:sediment = 9:1) (See Figure S7 for the detail). The partition coefficients of Sr and Nd during AOC dehydration were set to be 0.5 and 6.67, respectively (Kessel et al., 2005). The compositions of AOC are according to Staudigel et al. (1995). [c]B isotopes and concentrations of the detrital sediments (bulk) are according to Ishikawa and Nakamura (1993).

the southern Okinawa Trough (Figure 9). Recent studies have shown that Sr/Nd fractionation is negligible during sediment melting (Klaver et al., 2020; Martindale et al., 2013; Skora et al., 2015). Thus, the addition of sediment melts is indistinguishable with the addition of bulk sediments (Klaver et al., 2020).

AOC not only have high $^{87}Sr/^{86}Sr$ (up to ~0.707), but also relatively high concentrations of Sr (~180 ppm) (Staudigel et al., 1995), and, thus, AOC-derived fluids are most likely responsible for the decoupling of Sr-Nd isotopes. As illustrated in Figure 10, a mixture of fluids dehydrated from serpentinite (~20%) and AOC (~80%) can simultaneously increase the $\delta^{11}B$ values and $^{87}Sr/^{86}Sr$. Interestingly, some intra-oceanic arcs that are characterized by heavy B isotopes (e.g., Mariana Arc, South Sandwich Arc, and Tonga Arc) also show decoupled Sr-Nd isotopes, whereas their back-arc counterparts with low $\delta^{11}B$ show no Sr-Nd isotopes decoupling (Figure 11). Furthermore, a positive correlation has been observed between $\delta^{11}B$ and $^{87}Sr/^{86}Sr$ for arc lavas globally (De Hoog & Savov, 2018). This implies that serpentinite fluids are usually accompanied by AOC fluids when entering the source region of arc magmas. The mechanism, however, remains uncertain and requires further investigation. One possible explanation is that serpentinite fluids dehydrated from the slab mantle react with the overlying oceanic crust and, thus, inherit some compositional characteristics from the crustal materials (S. Chen, Hin, et al., 2019; Klaver et al., 2020).

5.4. Implications for Contribution of Multiply Sourced Subduction Components to Arc and Back-Arc Magmas

It is noted that both serpentinite fluids and AOC fluids are generally depleted in Nd (0.004 ppm and 1.1 ppm, respectively) compared to sediments (~16 ppm) (Table 1), thus these fluids are not likely to have contributed significant Nd to the mantle wedge. The relatively low $^{143}Nd/^{144}Nd$ of the Irabu Knoll lavas is most likely attributed to the addition of sediment Nd, probably in the form of bulk sediment (Shu et al., 2017) or a partial melt of sediment. Therefore, the Sr-Nd-B isotopic compositions of the Irabu Knoll lavas reflect a combination of contributions from subducted sediments, AOC fluids, and serpentinite fluids (Figure 10). Results of the mass balance modeling show that more than 90% of the total B budget of the Irabu Knoll lavas are from serpentinite and AOC fluids; AOC fluids also dominate the subduction input of Sr (>70%); whereas the Nd budget is mainly controlled by subducted sediments (>20%) and ambient mantle (>65%) (Figure 12).

The Yaeyama Graben lavas have the lowest $\delta^{11}B$ values (−3.7 ± 0.8‰), resembling those of the Mariana Trough lavas (−6.7‰ to −1.8‰; Chaussidon & Jambon, 1994; Chaussidon & Marty, 1995), Lau Basin lavas (−3‰ to −1.4‰; Chaussidon & Jambon, 1994), as well as the MORB (−7.1 ± 0.9‰; Marschall, 2018) (Figure 5). The Yonaguni Graben lavas have intermediate $\delta^{11}B$ values (+1.3 ± 0.4‰). Sr-Nd isotope decoupling is not observed in these relatively low-$\delta^{11}B$ back-arc lavas (Figure 4a), similar to the case of intra-oceanic back-arc lavas (Figure 11), suggesting no or very small amount of AOC fluids are incorporated into the back-arc mantle (Figure 10). Since the slab beneath the Yaeyama Graben (~110 km) is only slightly deeper than that beneath the Irabu Knoll (~90 km) (Figure 1), the sharp decrease of $\delta^{11}B$ values from the arc lavas to the back-arc lavas cannot be explained by slab dehydration induced isotope fractionation (Figure S7). It is also unlikely that the back-arc mantle is free of subduction components, as the lavas from the Yaeyama Graben and Yonaguni Graben still have elevated fluid mobile to fluid immobile element ratios relative to MORB

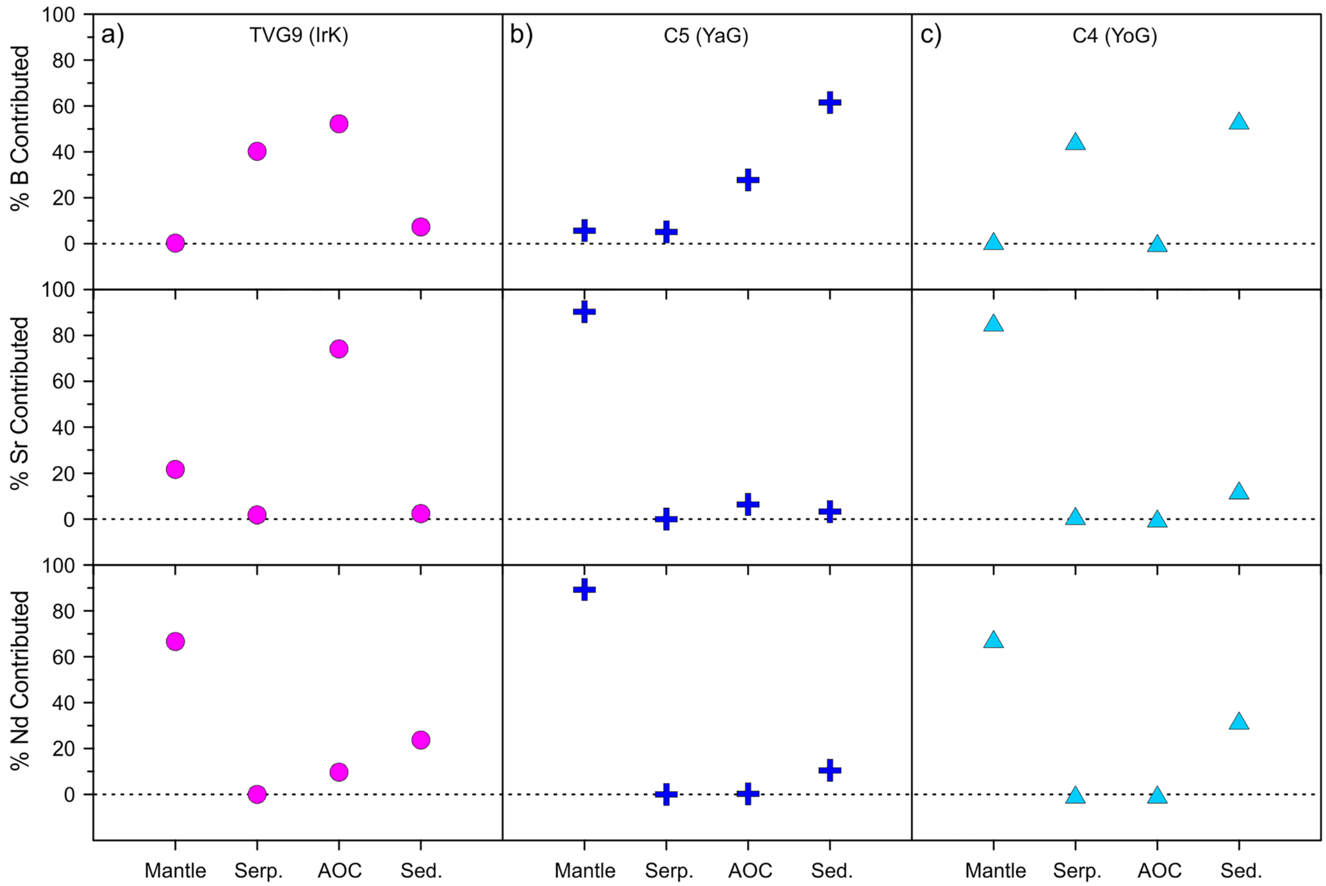

Figure 12. Results of the mass balance modeling showing the relative contributions of mantle, serpentinite fluids, AOC fluids, and sediments to the B, Sr, and Nd budget of the (a) Irabu Knoll (IrK), (b) Yaeyama Graben (YaG), and (c) Yonaguni Graben (YoG). The endmember compositions used for the modeling are given in Table 1. The results show that, with respect to subduction input, serpentinite fluids and AOC fluids are the major carriers of B; AOC fluids are the major carriers of Sr; sediments are the major carriers of Nd.

(Figure 6). Therefore, other types of subduction materials with low $\delta^{11}B$ are probably involved in the generation of the back-arc magmas. A recent study suggests that mantle wedge serpentinites ($\delta^{11}B = -14.5‰$ to +10‰) are potential fluid sources for low- to moderate-$\delta^{11}B$ arc magmas (Martin et al., 2020). For the Ryukyu subduction zone, however, the slab surface beneath the arc is too hot for any serpentine minerals to remain stable (Figure 9). Since the data of Yaeyama Graben and Yonaguni Graben mostly plot on the mixing lines between mantle and sediments (Figure 4), sediments are most likely the major subduction components added to the source regions of the back-arc magmas, which is consistent with the results of the mass balance modeling (Figure 12). The Yonaguni Graben lavas are also influenced by small amount of slab serpentinite fluids (Figures 10 and 12)

Our results also suggest that serpentinite fluids released from the subducting slab are only responsible for mantle melting over a limited area beneath the southern Okinawa Trough. This implies that migration of the serpentinite fluids through the mantle wedge does not occur by pervasive porous flow that would metasomatize a large portion of the mantle, but rather by a channelized transfer process in which the fluids have limited interaction with the surrounding mantle (Prigent et al., 2018; Tomanikova et al., 2019) or via a diapiric ascent of a mixture of slab and mantle lithologies (called "mélange") (Marschall & Schumacher, 2012; Nielsen & Marschall, 2017). Such focused flow has been considered to play a critical role in flux melting of sub-arc mantle (Saginor et al., 2013; Spandler & Pirard, 2013; Wilson et al., 2014). The concentrated or focused transport of material through the mantle wedge also allows multiply sourced components to contribute to the compositional heterogeneity of arc magmas.

6. Conclusions

We report the halogen (F and Cl) concentrations and Sr-Nd-Pb-B isotopic compositions of volcanic lavas from the Yaeyama Graben, Yonaguni Graben, and Irabu Knoll in order to constrain the influence of subduction components (AOC, sediments, and serpentinites) on the compositional heterogeneity of the lavas in the southern Okinawa Trough. The Irabu Knoll lavas have the heaviest B isotopes ($\delta^{11}B = +6.6 \pm 1.5‰$). Dehydration models suggest that fluids derived from AOC, sediments, and wedge serpentinites fail to explain the high $\delta^{11}B$ values of the Irabu Knoll lavas. Rather, we infer that they may be produced from sources that were influenced by slab-serpentinite-derived fluids, as has been proposed for the origin of heavy B isotopes in some intra-oceanic arc lavas (e.g., Mariana, Tonga, and South Sandwich). This indicates that B isotopes are still good tracers of serpentinite fluids for arc magmas that are largely affected by subducted sediments. The Yaeyama Graben lavas have much higher Cl concentrations and Cl/K, which is consistent with contamination by highly saline brines during magma ascent; whereas the progressive increase of F/Nd, the ratios of fluid mobile to fluid immobile elements, and $\delta^{11}B$ values from the Yaeyama Graben lavas to the Yonaguni Graben lavas, and to the Irabu Knoll lavas reflects an increasing input of fluids derived from serpentinite (±AOC).

We found that the arc lavas influenced by serpentinite fluids (with heavy B isotopes) generally show Sr-Nd isotopes decoupling, that is an increase in $^{87}Sr/^{86}Sr$ without significant changes in $^{143}Nd/^{144}Nd$ and, thus, deviate from the mantle-sediment mixing trend, which is a common observation in arcs. The heavy B isotopes plus decoupled Sr-Nd isotopes cannot be explained by serpentinite fluids alone, but can be well accounted for by a combination of fluids from serpentinite and AOC. Therefore, we suggest that recycling of serpentinite fluids and AOC fluids in subduction zones are usually coupled. We speculate that the large variation of Sr-Nd-B isotopes in the southern Okinawa Trough lavas reflects the contributions of multiply sourced subduction components (e.g., serpentinites, AOC, and sediments): serpentinite fluids and AOC fluids are the major carriers of B; AOC fluids dominate the input of Sr; Nd is mainly carried by sediments. These components can migrate through the mantle wedge via a channelized flow and play a significant role in the chemical heterogeneity of arc magmas.

Data Availability Statement

All the data that support the findings of this study are given in supporting information and are also deposited in Mendeley data set (http://doi.org/10.17632/mw54t5wt68.1).

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