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Lithium, Oxygen and Magnesium Isotope Systematics of Volcanic Rocks in the Okinawa Trough: Implications for Plate Subduction Studies

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Abstract: Determining the influence of subduction input on back-arc basin magmatism is important for understanding material transfer and circulation in subduction zones. Although the mantle source of Okinawa Trough (OT) magmas is widely accepted to be modified by subducted components, the role of slab-derived fluids is poorly defined. Here, major element, trace element, and Li, O and Mg isotopic compositions of volcanic lavas from the middle OT (MOT) and southern OT (SOT) were analyzed. Compared with the MOT volcanic lavas, the T9-1 basaltic andesite from the SOT exhibited positive Pb anomalies, significantly lower Nd/Pb and Ce/Pb ratios, and higher Ba/La ratios, indicating that subducted sedimentary components affected SOT magma compositions. The δ^{7} Li, δ^{18} O, and δ^{26} Mg values of the SOT basaltic andesite (-5.05% to 4.98%, 4.83% to 5.80% and -0.16‰ to -0.09‰, respectively) differed from those of MOT volcanic lavas. Hence, the effect of the Philippine Sea Plate subduction component, (low δ^7 Li and δ^{18} O and high δ^{26} Mg) on magmas in the SOT was clearer than that in the MOT. This contrast likely appears because the amounts of fluids and/or melts derived from altered oceanic crust (AOC, lower δ^{18} O) and/or subducted sediment (lower δ^7 Li, higher δ^{18} O and δ^{26} Mg) injected into magmas in the SOT are larger than those in the MOT and because the injection ratio between subducted AOC and sediment is always >1 in the OT. The distance between the subducting slab and overlying magma may play a significant role in controlling the differences in subduction components injected into magmas between the MOT and SOT.

Keywords: Li-O-Mg isotopes; magma; plate subduction; Okinawa Trough; western Pacific

1. Introduction

The Okinawa Trough is a young back-arc basin in the western Pacific, and its magmas have been affected by the subducted Philippine Sea plate (PSP) [1]. The chemical and stable isotope (Li, O, and Mg) compositions of back-arc volcanic lavas are conventionally used to study the contributions of subducted slabs to magmas [1–11]. However, magmas produced by plate subduction are relatively enriched in large ion lithophile elements and light rare earth elements, but are depleted in high field strength elements; these magmas usually have low Ce/Pb ratios since Pb is preferentially extracted from subducted oceanic crust and/or sediments during plate subduction and dehydration processes [8–10,12]. As incompatible elements share similar partition coefficients in magmas, their ratios are rarely modified by the partial melting or crystallization processes. Thus, the incompatible element ratios can be used to trace the compositions of mantle sources. For example, Ba/La

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ratio is a good indicator of subduction components [13]. Ce/Pb ratios of sediments exhibit a distinctive range, below the values in mantle rocks, and are not significantly affected by fractional crystallization and partial melting processes [14]; thus, these ratios can be used to estimate the contributions of plate subduction components. Th concentrations of marine sediments are higher than the mantle values, and Th is fluid immobile relative to large ion lithophile elements. A high Th/Rb ratio in magma indicates that a sediment melt was added to the magma source [15]. In addition, high Th/La, Th/Nb, and Th/Nd ratios of the volcanic rocks indicate the contribution of sediment melts to the mantle wedge [16– 19]. Ba/La, Cs/La, B/Nb, Pb/Ce, B/La, and Li/Y ratios can also serve as proxies for fluids dehydrated from subducted sediments [20].

Moreover, lithium (Li) is a fluid mobile element, and considerable Li isotope fractionation occurs during oceanic crust alteration and plate subduction [21-24]. Subducted sediments and altered oceanic crust (AOC) have high Li contents (up to 80 ppm) and a broad range of δ^7 Li values (-12‰ to +21‰) [4,25–28]. During the metamorphism and dehydration of subducted slabs, fluids carrying heavy Li isotopes are released from the AOC and metasomatize the overlying mantle, resulting in a high- δ^7 Li mantle wedge and a low- δ^7 Li residual plate [29–31]. However, Guo, et al. [1] studied Li isotope data from middle Okinawa Trough (MOT) volcanic lavas and found that the MOT magmas were affected by subduction components (with AOC:sediment = 96:4). The δ^7 Li values of volcanic lavas from the Izu Arc in Japan vary across the arc, decreasing with increasing depth to the subducting plate, indicating that the amount of slab fluids decreases with increasing depth [32–34]. Furthermore, Benton, et al. [35] analyzed the Li isotope compositions of volcanic lavas in the Mariana Arc front seamounts and found that they have heterogeneous δ^7 Li signatures, which reflect a complex history of exchange between the slab fluids and the forearc mantle. These results indicate that there may be uncertainties in using Li isotope data for back-arc volcanic lavas to uncover magma sources and plate subduction and implies that other isotopic tools should be used to clarify magmatic processes in western Pacific plate subduction zones.

However, the oxygen (O) isotopic compositions of the MOT volcanic lavas reveal that the magma source is mantle peridotites modified by subducted slab components [36]. The O isotopic compositions of volcanic glasses obtained from the Manus Basin and the Mariana Trough indicate that the Mariana Trough magma was affected by subduction-modified mantle [37] and that the Manus Basin magma had a δ^{18} O-depleted mantle reservoir, wherein δ^{18} O was increased by recent subduction and a sediment component with low δ^{18} O and high ³He/⁴He values was derived from the Manus Basin plume [37]. Moreover, the O isotope compositions of volcanic glasses and phenocrysts from the Lau Basin suggest that a subduction cycle involving oxygen-rich mantle materials altered the original δ^{18} O of the magma [38]. All these results indicate that the O isotope data for volcanic lavas may not completely reflect plate subduction processes.

Magnesium (Mg) isotopes are important geochemical tracers of both magmatic sources containing recycled crustal materials [39–41] and sources of fluids dehydrated from hydrous minerals in subducting plate [5,40,42–44]. However, the Mg isotope compositions of white schist in the western Alps reveal that the dehydration of serpentinites formed in the mantle wedge during plate subduction and exhumation can release Mgrich fluids to the subduction channel [5]. Furthermore, a study of the Mg isotope compositions of prograde metamorphic rocks from eastern China revealed that Mg isotope fractionation was limited during continental subduction [41]. All these results also indicate that the Mg isotope data for volcanic lavas may not completely determine magma sources and plate subduction processes.

Therefore, we sought to combine the Li isotope composition of volcanic lavas with their O and Mg isotope compositions to understand the magmatic processes involved in forming volcanic lavas and the influence of plate subduction on magmas in the Okinawa Trough (OT). However, previous studies have shown that subducted sediments significantly influence the mantle source of magmas beneath the OT [2,10,19,45,46]. Although

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slab-derived fluids are also considered to contribute to the OT magmas [47–50], the evidence for this contribution is insufficient. Furthermore, Li and O isotope studies have been conducted in the OT, but the data remain limited to only three Li isotope analyses in the MOT. O isotope studies have primarily focused on crustal contamination as a source of magma formation [36,51–53], and the study of the Mg isotopic compositions of volcanic lavas has not yet been applied to the OT. Here, we investigate the major and trace elements as well as the Sr, Nd, Li, O, and Mg isotopic compositions of volcanic lavas from the MOT and SOT, with the goal of establishing how OT volcanic lavas form and what they represent for understanding the effects of subduction components on back-arc magmas.

2. Geologic Setting

The OT is a young back-arc basin developed in response to the subduction of the PSP (Figure 1) [11,54–56] and provides a window into understanding the influences of plate subduction on the back-arc basin evolution, mantle melting, crust-mantle interactions, and seafloor hydrothermal activity [1,11,46,57,58]. According to the prevailing tectonics, the OT can be divided into three parts: northern OT (NOT), MOT, and SOT, bounded by the Tokara Fault and the Kerama Fault, respectively [1,11,50]. The crustal thicknesses of the NOT, MOT, and SOT segments are 15 to 23 km, 12 to 18 km, and 10 to 16 km, respectively [54,59–64]. The depths to the Mohorovičić discontinuity (Moho) are 27–30 km and 16–22 km in the NOT and SOT, respectively [59,65,66]. The subduction direction of PSP is nearly perpendicular to the axis of the MOT, with a slab depth of 150–200 km, and becomes progressively more oblique to the south, with a slab depth of ~150 km [11,47,50,67,68]. The subduction rates of PSP increase from 4.9 to 7.3 cm/a from north to south [11,56]. In addition, the convergence rate gradually increases from ~6–7 cm/a in central Ryukyu to ~7–13 cm/a in southern Ryukyu [11,56,69], and the back-arc extension rate increases from 2 cm/a in the NOT to 5 cm/a in the SOT [65,69,70].



Figure 1. Regional geologic map of the Okinawa Trough (OT) showing the sampling locations. MOT, middle Okinawa Trough; SOT, southern Okinawa Trough; and NOT, northern Okinawa Trough. Black solid lines represent the depth contours of the subducting plate (Wadati-Benioff zone) [11,67,71]. Yellow dotted lines mark the Tokara Fault and the Kerama Fault.

The OT has undergone substantial magmatic activity that has generated abundant volcanic lavas. The NOT is dominated by rhyolites and dacites [6,53]. The magma in the NOT could be generated by the differentiation of basaltic melt contaminated by an enriched crustal component [53]. The MOT volcanic lavas exhibit bimodal compositional distribution, forming a basaltic-rhyolitic dominant suite with scarce andesitic lavas [6,72,73]. The Sr-Nd-Pb isotope compositions suggest that the magma source of the MOT

volcanic lavas resembles the depleted mantle (DM) but also shows signatures of the enriched mantle II (EMII), indicating that the mantle source for the MOT volcanic lavas is a hybrid of the DM and EMII endmembers [74]. Geochemical modelling shows that the magma source for the MOT volcanic lavas could be generated by hybridizing approximately 0.8-2.0% subducted sediments with 98.0-99.2% mantle rocks [74]. The SOT is dominated by rhyolites, basalts and basaltic andesites [50,72,75,76]. In the SOT, the petrogenesis of rhyolites is due to the ascent of basaltic magma from a deep magma reservoir to a shallow depth where fractional crystallization and crustal assimilation occurred [75]. The decrease in the DUPAL-like anomaly from the rhyolites sampled in the western part of the MOT to the volcanic lavas in the MOT axial zone can be explained by the injection of asthenospheric mantle into the pre-existing mantle with DUPAL-like signature during back-arc extension. The Tl-Pb-Sr-Nd isotopic compositions of volcanic lavas from the NOT to the SOT can be accounted for by the sediment inputs of <1%, 0.1-1% and 0.3-2%, respectively, by weight to the depleted mantle source [46]. The magma sources of the SOT basalts are mainly affected by slab fluids and bulk sediments [47]. The magma sources of the MOT basalts are affected by fluids derived from both sediment and AOC [47]. Based on the above findings, we aim to use the geochemical and Li-O-Mg isotope compositions of OT volcanic lavas to understand the origins of magmas and the effects of plate subduction on magmas in this study.

3. Sampling and Methods

3.1. Sample Collection

The volcanic lavas were collected from the MOT and SOT using a TV grab sampler in 2014 and 2016 during HOBAB (Hello Back Arc Basin) 2 and 4 cruises, respectively. The R2, T5-2, and T2 samples were obtained from the Iheya Ridge (MOT), samples T6-1 and T6-2 were obtained from the western slope of the Iheya Ridge in the MOT, and sample T9-1 was obtained from the Yaeyama Graben (SOT) (Table S1 and Figure 1).

T9-1 basaltic andesite from the SOT is a black vesicular lava sample with a compact, massive structure. It contains few phenocrysts, which predominantly consist of clinopyroxene (~5%), orthopyroxene (~5%), plagioclase (~10%), a small amount of olivine (~1%), and accessory minerals (magnetite and ilmenite). The groundmass of the T9-1 basaltic andesite is mostly composed of plagioclase and clinopyroxene microlites (Figure 2a). The phenocrysts in the T9-1 basaltic andesite are euhedral to subhedral and range from ~0.1 to ~1.0 mm in size.



Figure 2. Representative photomicrographs of the MOT and SOT volcanic lava samples. (**a**) T9-1 is from the SOT and (**b**) R2, (**c**) T5-2, (**d**) T2, (**e**) T6-1, and (**f**) T6-2 are from the MOT. Abbreviations: olivine (Ol); magnetite (Mt); clinopyroxene (Cpx); orthopyroxene (Opx); and plagioclase (Pl).

R2 basalt from the MOT is a black lava with a moderately porphyritic massive structure. It contains olivine (~5%), plagioclase (~5%), and clinopyroxene (~1%) phenocrysts. The phenocrysts in the R2 basalt are euhedral to subhedral, with sizes ranging from ~0.05 to 6 mm. The groundmass of the R2 basalt is dominated by clinopyroxene and plagioclase microlites (Figure 2b). Both the T5-2 trachyandesite and T2 andesite from the MOT are black lavas with compact, massive structures. Thin sections reveal that the T5-2 trachyandesite and T2 andesite are porphyritic and that ~15% of the phenocrysts are plagioclase with minor clinopyroxene and orthopyroxene. The phenocrysts in the T5-2 trachyandesite and T2 andesite samples are euhedral to subhedral, with sizes ranging from ~40 to ~300 μ m (Figure 2c,d). The T6-1 pumice from the MOT (Figure 2e) is a light, white, vesicular lava. The phenocrysts (<5%) in the T6-1 pumice are primarily clinopyroxene, orthopyroxene, and plagioclase and range from ~0.2 to 0.5 mm in size. The groundmass in the T6-1 pumice is mainly composed of plagioclase microlites (Figure 2e). The T6-2 pumice from the MOT is a gray-black vesicular lava. The phenocrysts (<5%) in the T6-2 pumice mainly consist of clinopyroxene, plagioclase, and orthopyroxene, with sizes ranging from ~0.2 to 0.4 mm in size. The groundmass in the T6-2 pumice is also mainly composed of plagioclase microlites (Figure 2f).

Details about the sample processing, electron microprobe, major element and trace element, and Sr and Nd isotope analytic methods are given in the Supplementary Texts, and the sampling locations and sample analytic results can be found in Tables S1–S8.

3.2. Li, O, and Mg Isotope Analyses

The whole-rock powders and minerals were fully digested. All reagents (HF, HNO₃, and HCl) were doubly distilled, and Milli-Q[®] water (18.2 M Ω ·cm) (Merck Millipore Inc., Billerica, MA, USA) was used. For the isotopic analyses, Li was separated using organic solvent-free two-step liquid chromatography in a clean laboratory at the University of Science and Technology of China (USTC). The procedure used was described by [77]. All separations were monitored using inductively coupled plasma mass spectrometry (ICP-MS) analysis to guarantee both a high Li yield (>99.8% recovery) and a low Na/Li ratio (<0.5). The final Li concentrations of the solutions used for the multi-collector ICP-MS (MC-ICP-MS) analyses were targeted in the range of 50–100 ppb to ensure the precision and accuracy of the results. The total procedural blanks for column chromatography alone and for sample digestion and column chromatography combined were <~0.03 ng of Li. Compared with the ~200–5000 ng of Li used for our analyses, the blank correction was negligible at the uncertainty levels achieved (≤0.2 ‰, see below).

The lithium isotope compositions were analyzed at the USTC using a Neptune Plus MC-ICP-MS system (Thermo Fisher Scientific Inc., Waltham, MA, USA)) operating in wet plasma mode using an X skimmer cone and jet sample cones (Table S9). The samples were introduced through a low-flow PFA nebulizer (~50 µL/min) coupled with a quartz spray chamber. The ⁷Li and ⁶Li isotopes were measured simultaneously by two opposing Faraday cups. Each sample analysis was bracketed before and after by analyses of the reference material L-SVEC. For a solution containing 100 ppb of Li and an uptake rate of 50 µL/min, the typical ⁷Li intensity was ~8 V. The in-run precision of the ⁷Li/⁶Li measurements was ≤0.2‰ for one block of 60 ratios. The external precision was based on the long-term analysis of an in-house standard (Li-QCUSTC = +8.8‰ ± 0.2‰ (2 standard deviations (SD), *n* = 161)). For international rock standards, repeat analyses at the USTC yielded values of +4.4‰ ± 0.3‰ (2SD, *n* = 8) for BHVO-2, -0.8‰ ± 0.3‰ (2SD, *n* = 29) for GSP-2, and +5.9‰ ± 0.5‰ (2SD, *n* = 9) for AGV-1, which were within the uncertainty limits of previously published results [77,78]. The results are reported in the delta notation [δ ⁷Li = ((⁷Li/⁶Li)_{sample}/(⁷Li/⁶Li)_{standard} -1)) × 1000] relative to the L-SVEC Li isotope standard [79].

The whole-rock and mineral oxygen isotope compositions were measured using laser fluorination with a 25 W MIR-10 carbon dioxide (CO₂) laser at the CAS Key Laboratory of Crust-Mantle Materials and Environments at the USTC, Hefei. The oxygen isotope analyses and the data acquisition followed the methods described by [80,81]. O₂ was extracted from the samples through reaction with bromine pentafluoride (BrF₅) in nickel (Ni) bombs, and it was then converted to CO₂ through reaction with a hot carbon rod. The δ^{18} O of CO₂ was measured using a Delta+ mass spectrometer. Reference minerals GBW04409 quartz (δ^{18} O = 11.11‰ ± 0.1‰) [81] and in-house standard 04BXL07 garnet (δ^{18} Ov-smow = $3.70\% \pm 0.1\%$) [82] were analyzed during each run. We analyzed the O isotope compositions of the whole rocks and the minerals from each sample twice, except for R2-2-Cpx and T2-Opx (Table S9). The data are reported using the usual δ^{18} O notation relative to Vienna standard mean ocean water (V-SMOW). On a given day, the reproducibility of each standard was better than $\pm 0.2\%$ (2 σ) for δ^{18} O.

The Mg isotopes were measured using a Thermo Scientific Neptune Plus MC-ICP-MS system following the methods of [83] at the CAS Laboratory of Crust-Mantle Materials and Environments at the USTC, Hefei. The whole-rock powders and minerals were fully digested to obtain ~20 µg of Mg for chemical purification. A mixture of concentrated HF-HNO3 was used for digestion. Mg was purified in Savillex columns loaded with 2 mL of Bio-Rad AG[®]50W-X12 resin (Bio-Rad Laboratories Inc., Hercules, CA, USA). For the Mg isotope analyses, sample-standard bracketing was used, wherein DSM-3 was the bracketing standard. The Mg isotope compositions are reported using the standard δ notation relative to DSM-3. The uncertainties in the δ^{25} Mg and δ^{26} Mg values of the standards and the samples are reported as 2SD based on repeated measurements (Table S9). The data quality was carefully controlled through the repeated analysis of multiple Mg isotope standards (Table S9). The long-term external precision was better than ±0.05‰. Furthermore, each sample was processed three times under the same conditions. During the analyses, the δ^{26} Mg values obtained for BCR-2 (-0.215‰ ± 0.017‰; n = 3) and BHVO-2 $(-0.205\% \pm 0.020\%; n = 3)$ were all identical within the established uncertainty limits (-0.162‰ ± 0.014‰ for BCR-2 and -0.216‰ ± 0.035‰ for BHVO-2) [83].

4. Results

4.1. Clinopyroxene and Orthopyroxene

The clinopyroxene contents in R2 basalt, the orthopyroxene contents in T5-2 trachyandesite, T2 and site, T6-1 and T6-2 rhyolites from the MOT, and the orthopyroxene content in T9-1 basaltic and site from the SOT, were analyzed using an electron microprobe. The data are presented in Tables S2–S6. The clinopyroxene and orthopyroxene were unzoned, and their compositions were homogeneous (Figure 2 and Tables S2–S6). There was no notable difference between the Mg#s of clinopyroxene and orthopyroxene [80–84 in R2 (n = 11), 67–76 in T9-1 (n = 18), 59–72 in T2 (n = 14), 34–48 in T6-1 (n = 29), and 47–52 in T6-2 (n = 6)] (Tables S2–S6).

However, assuming that the mineral phases represent equilibrium compositions, the OT clinopyroxene and orthopyroxene composition data (Tables S2–S6) allowed the use of the clinopyroxene and orthopyroxene-liquid geothermobarometer of [84,85] to calculate the crystallization temperatures and pressures of the magma along its ascent path. The standard errors of the temperature and pressure calibrations were 39 °C and 2.1 kbar, respectively [85]. According to the clinopyroxene-liquid geothermobarometer (Table S4) [84], the crystallization temperature, pressure, and magma depth for formation of the clinopyroxene phenocrysts in the R2 basalt from the MOT were estimated to be in the ranges of 1112–1137 °C, 0.14–0.42 GPa, and ~8.0–16.9 km, respectively. From the orthopyroxeneliquid geothermobarometers (Table S3) [85], the crystallization temperature, pressure, and magma depth for formation of the orthopyroxene phenocrysts in the T2 and esite from the MOT were calculated to be in the ranges of 1021–1095 °C, 0.03–0.37 GPa, and 4.3–15.3 km, respectively (Table S3). The crystallization temperature, pressure, and magma depth for formation of the orthopyroxene phenocrysts in the T6-1 and T6-2 rhyolites from the MOT were estimated to be 821-884 °C and 844-858 °C, 0.11-0.59 GPa and 0.01-0.11 GPa, and 7.1–22.2 km and 3.7–7.1 km, respectively (Tables S5 and S6). The crystallization temperature, pressure, and magma source depth for forming the orthopyroxene phenocrysts in the T9-1 basaltic andesite from the SOT were estimated to be 1095–1138 °C, 0.16–0.40 GPa, and 8.4–16.1 km, respectively (Table S2).

4.2. Major Element and Trace Element Compositions of the Volcanic Lavas

The major element concentrations of the volcanic lavas obtained from the MOT and SOT are presented in Table S7. No increase in ⁸⁷Sr/⁸⁶Sr ratios was observed with increasing LOI (Figure S1), indicating that the samples were free of seawater alteration. On the total alkalis-silica (TAS) diagram, the volcanic lavas plot in the R2 basalt, T9-1 basaltic andesite, T5-2 trachyandesite, T2 andesite, T6-1 and T6-2 rhyolite fields, indicate they can be classified as subalkaline (SiO₂ = 51.62 – 73.85 wt.%; Na₂O + K₂O = 2.63 – 8.16 wt.%) (Figure 3) [86–88]. On the K₂O vs. SiO₂ diagram, the T9-1 basaltic andesite from the SOT and the R2 basalt from the MOT plot in the low-K arc tholeiitic field, the T5-2 trachyandesite and T2 andesite from the MOT plot in the medium-K calc-alkaline field, and the T6-1 and T6-2 rhyolites from the MOT, are in the high-K calc-alkaline field (Figure 3). On the Harker diagrams (Figure S2), the major element oxides (except for Na₂O, TiO₂, and P₂O₅) of the MOT volcanic lavas exhibit first-order trends, indicating that they evolved through the fractional crystallization of magma [89]. In addition, the T9-1 basaltic andesite from the MOT volcanic lavas (Na₂O = 2.77–5.43 wt.%, Al₂O₃ = 12.23–17.08 wt.%) (Figure S2).



Figure 3. Classification diagrams for the MOT and SOT volcanic lavas. (**a**) Plots of Na₂O + K₂O vs. SiO₂. The base diagram is from [87], and the boundary between the alkaline and subalkaline rocks is from [86]. (**b**) K₂O (wt.%) vs. SiO₂ (wt.%). Boundaries are from [88]. Data for published mafic samples are from [1,36,50,74]. Data for published felsic samples from [1,73,74]. Data for samples R2, T5-2, T6-1, T2, T6-2, and T9-1 are from this study.

The trace element concentrations of the OT lava samples are presented in Table S8. The primitive mantle-normalized spider diagrams for the MOT and SOT volcanic lavas reveal obvious enrichment in large ion lithophile elements relative to the high field strength elements and rare earth elements (Figure 4). All the samples had negative niobium (Nb), tantalum (Ta), and titanium (Ti) anomalies and distinctly positive Pb anomalies (Figure 4). Moreover, the MOT rhyolites exhibited significant Sr, P, and Ti depletions (Figure 4e,f), suggesting mineral fractionation. The R2 basalt from the MOT and the T9-1 basaltic andesite from the SOT had slight Sr enrichments (Figure 4a). The chondrite-normalized REE diagrams of the MOT and SOT volcanic lavas reveal light REE (LREE) enrichment relative to heavy REEs (HREEs) (Figure 4), and the MOT volcanic lava samples ((La/Yb)_N = 2.23–3.68) were more fractionated than the T9-1 basaltic andesite sample ((La/Yb)_N = 1.66) from the SOT. The MOT rhyolites exhibited significant negative europium (Eu) anomalies (Eu/Eu^{*}T6-1 = 0.55, and Eu/Eu^{*}T6-2 = 0.36, where Eu/Eu^{*} = 2Eu_N/(Sm_N + Gd_N)), the MOT trachyandesite and andesite exhibited small negative Eu anomalies

 $(Eu/Eu^*_{T5-2} = 0.85, and Eu/Eu^*_{T2} = 0.86)$, and the MOT basalt and the SOT basaltic andesite exhibited negligible Eu anomalies (Eu/Eu^*_{R2} = 0.99, and Eu/Eu^*_{T9-1} = 1.05). However, fractionation between the LREEs and HREEs was higher in the MOT volcanic lavas ((La/Yb)N = 2.23–3.68) than in the SOT lava ((La/Yb)N = 1.66).



Figure 4. (**a**,**c**,**e**) Trace element patterns normalized to primitive mantle and (**b**,**d**,**f**) corresponding rare earth element (REE) patterns normalized to chondritic contents for the MOT and SOT volcanic lavas. Primitive mantle and chondrite contents are from [90]. (**a**,**b**) show the trace element distributions of the MOT basalt and the SOT basaltic andesite, respectively. (**c**,**d**) show the trace element distributions of the MOT andesite and trachyandesite, respectively. (**e**,**f**) show the trace element distributions of the MOT rhyolites. Data are from [50,73], and this study.

4.3. Li–O–Mg Isotope Compositions of the Volcanic Lavas and Minerals

The Li isotopic compositions of the OT volcanic lavas and minerals are reported in Table S9 and are plotted in Figure 5a,b. The δ^7 Li values of all samples vary from -5.05%to +5.61‰ (Figure 5a). The highly fractionated rhyolites (T6-1 and T6-2) have higher δ ⁷Li values than the mafic and intermediate lavas (Figure S3b), probably as a result of latestage magma evolution; thus, these data are not included in the following discussion. The T5-2 trachyandesite from the MOT had lower δ^7 Li values (-0.83‰ to +2.90‰) than the other MOT samples (Figure 5a). The majority of the 87Li values in the MOT volcanic lavas fell within the range of mid-ocean ridge basalts (MORBs) (+1.50% to +6.85%) (Figure 5a,b) [22,33,91–93], indicating that these samples originated from similar magmatic sources. The glass in T9-1 basaltic and esite from the SOT had the highest δ^{7} Li value (+4.98‰) (Table S9), while the δ^7 Li values of clinopyroxene (+0.49‰), orthopyroxene (+1.04%), and plagioclase (-5.05%) in the T9-1 basaltic and esite sample from the SOT were all lower than those of MORBs (Table S9 and Figure 5a). The δ^{7} Li values of the MOT and SOT plagioclase and clinopyroxene phenocrysts were lower than those of olivine, orthopyroxene, and glass (Table S9 and Figure 5a). Furthermore, most of the δ^7 Li values obtained in this study were lower than those obtained for the MOT volcanic lavas by [1] $(\delta^{7}\text{Li} = +2.6\% \text{ to } +6.9\%, n = 3)$. The $\delta^{7}\text{Li}$ values of the OT basaltic lavas $(\delta^{7}\text{Li}_{R2-1} = +3.33\%,$ and δ^7 Li₉₋₁ = +3.45‰) were lower than those reported for the Lau Basin basalts (δ^7 Li = +4.32‰ to +4.82‰) [4].



Figure 5. The δ^7 Li distributions of (**a**) the different whole-rock and mineral separates from the Okinawa Trough (OT) and (**b**) mid-ocean ridge basalts (MORBs), AOC, sediment, upper continental crust, and seawater. The δ^{16} O distributions of (**c**) the different whole-rock and mineral separates from the OT and (**d**) MORBs, AOC, sediment, and upper continental crust. The δ^{26} Mg distributions of (**e**) the different whole-rock and mineral separates from the OT and (**f**) MORBs, AOC, sediment, upper continental crust, and seawater. The MORB δ^7 Li range is from [22,33,91–93]. The AOC δ^7 Li data are from [4,22,25,94]. The sediment δ^7 Li data are from [3,4,25,27,95–97]. Upper continental crust

(UCC) δ^7 Li data are from [98]. The MORB δ^{18} O range is from [99–101]. AOC δ^{18} O data are from [102,103]. Sediment δ^{18} O data are from [104]. UCC δ^{18} O data are from [105]. The MORB δ^{26} Mg range is from [106,107]. AOC δ^{26} Mg data are from [39]. Sediment δ^{26} Mg data are from [108]. UCC δ^{26} Mg data are from [109].

In addition, the Li isotope fractionations ($\Delta^7 \text{Li}_{x-y} = \delta^7 \text{Li}_x - \delta^7 \text{Li}_y$) between coexisting mineral phases (*x*-*y*) were also calculated. The $\Delta^7 \text{Li}_{cpx-ol}$ (Li isotope fractionation between coexisting clinopyroxene and olivine) and $\Delta^7 \text{Li}_{pl-ol}$ (Li isotope fractionation between coexisting plagioclase and olivine) values of the R2 basalt from the MOT were –3.76 and –3.77, respectively. The $\Delta^7 \text{Li}_{cpx-ol}$, $\Delta^7 \text{Li}_{opx-ol}$, and $\Delta^7 \text{Li}_{pl-ol}$ values of the T5-2 trachyandesite from the MOT were –3.24, –0.07, and –0.55, respectively. The $\Delta^7 \text{Li}_{cpx-ol}$, $\Delta^7 \text{Li}_{opx-ol}$, $\Delta^7 \text{Li}_{pl-ol}$ values of the T9-1 basaltic andesite from the SOT were –2.35, –1.80, and –7.88, respectively. The $\Delta^7 \text{Li}$ values of all samples were negative. This cannot be explained by closed system equilibrium between the different minerals and olivine.

The oxygen isotopic compositions of the OT volcanic lavas and minerals varied from +4.83% to +6.69% (Table S9 and Figure 5c). The δ^{18} O values showed no significant increase with increasing SiO₂ concentrations for whole-rock samples (Figure S3a). The δ^{18} O values (+5.38% to +5.79%) of R2-1 and R2-2 basalts from the MOT fell within the MORB range (Figure 5c,d) (+5.30% to +5.80%) [110,111]. The δ^{18} O values of the T5-2 trachyandesite exhibited a broader range (+5.15‰ to +6.69‰) than those of the other MOT samples (Figure 5c). The majority of the δ^{18} O values (+4.83‰ to +5.80‰) of the T9-1 basaltic and esite and its mineral samples from the SOT were lower than those of the MOT volcanic lava samples (+5.15‰ to +6.69‰) (Table S9 and Figure 5c). Moreover, the δ^{18} O values of all the OT volcanic lava and mineral samples analyzed in this study overlapped with the δ^{18} O range previously reported for volcanic lavas from the Manus Basin (+4.96‰ to +6.68‰) [37,100]. The whole-rock δ^{18} O values of T5-2 (+6.69‰) and T6-2 (+6.08‰) fell within the range of the MOT volcanic lavas (+6.0‰ to +7.6‰) analyzed by [36]. The δ^{18} O values of the T9-1 (+5.29‰), R2-1 (+5.41‰), R2-2 (+5.38‰), T2 (+5.69‰), and T6-1 (+5.84‰) wholerock samples (Table S9) were lower than those of the volcanic lavas (+6.6‰ to +8.8‰) analyzed by [36,53]. The majority of the δ^{18} O values of the T9-1 basaltic and esite from the SOT and the R2-1 and R2-2 basalts from the MOT were lower than those measured for the volcanic lavas of the North Fiji Basin ($\delta^{18}O = +5.78\%$ to +6.06%) [38] and the Mariana back-arc basin ($\delta^{18}O = +5.8\%$ to +6.0‰) [7].

In addition, the δ^{18} O values of plagioclase and glass were higher than those of olivine, clinopyroxene, and orthopyroxene in the MOT and SOT volcanic lavas (Table S9), and the δ^{18} O values of olivine, clinopyroxene, and orthopyroxene exhibited smaller variations than those of volcanic glass (Figure 5c). The $\Delta^{18}O_{cpx-ol}$ and $\Delta^{18}O_{pl-ol}$ values calculated for the R2 basalt from the MOT were 0.08 and 0.22, respectively. The $\Delta^{18}O_{cpx-ol}$, $\Delta^{18}O_{opx-ol}$, and $\Delta^{18}O_{pl-ol}$ values calculated for the T5-2 trachyandesite from the MOT were -0.09, 0.41, and 0.75, respectively. The $\Delta^{18}O_{cpx-ol}$, $\Delta^{18}O_{opx-ol}$, and $\Delta^{18}O_{pl-ol}$ values calculated for the T9-1 basaltic andesite from the SOT were 0.16, 0.67, and 0.66, respectively. These data imply that the crystallization of the magma and minerals preferentially incorporated isotopically heavy O into the orthopyroxene and plagioclase phenocrysts, resulting in the relative enrichment of lighter O isotope in the olivine phenocrysts.

The Mg isotope compositions of the OT volcanic lavas and their minerals varied from -0.31% to -0.09% (Table S9 and Figure 5e). No correlations existed between Mg isotopes and SiO₂ concentrations for whole-rock samples (Figure S3c). The δ^{26} Mg values of the R2 basalt from the MOT ranged from -0.31 to -0.20%, and all the values fell within the MORB range ($-0.26\% \pm 0.07\%$) [107] (Figure 5e,f and Table S9). The whole-rock δ^{26} Mg values (-0.25% to -0.11%) of the MOT andesite (T2) and trachyandesite (T5-2) were slightly higher than those of MORBs. All the δ^{26} Mg values (-0.16% to -0.09%) of the T9-1 basaltic andesite from the SOT were higher than those of MORBs (Figure 5e,f). The δ^{26} Mg values of the clinopyroxene, orthopyroxene, and glass in the T9-1 basaltic andesite from the SOT were higher than those for those from the MOT volcanic lavas (Table S9 and

Figure 5e). Furthermore, the $\Delta^{26}Mg_{cpx-ol}$ value calculated for the R2 basalt from the MOT was -0.01. The $\Delta^{26}Mg_{cpx-ol}$ and $\Delta^{26}Mg_{opx-ol}$ values calculated for the T5-2 trachyandesite from the MOT were -0.01 and -0.02, respectively. The $\Delta^{26}Mg_{cpx-ol}$ and $\Delta^{26}Mg_{opx-ol}$ values calculated for the T9-1 basaltic andesite from the SOT were 0.02 and 0.01, respectively.

However, only the δ^7 Li (+3.33‰ to +4.09‰), δ^{18} O (+5.41‰ to +5.78‰), and δ^{26} Mg (-0.28‰ to -0.20‰) values of the glass or whole rock from the mafic R2 basalt might be considered indicative of magma source compositions.

5. Discussion

5.1. Genesis and Evolution of Magmas

The crystallization temperatures and pressure ranges (Tables S2–S6) of the corresponding ascending magmas were estimated from the pyroxene compositions of the volcanic lavas (Section 4.1) [84,85,112,113]. The crystallization temperature, pressure, and magma depth for forming the orthopyroxene phenocrysts in T6-2 rhyolites from the MOT were lower and shallower than those for forming the clinopyroxene phenocrysts in the R2 basalt, the orthopyroxene phenocrysts in the T2 andesite from the MOT, and the orthopyroxene phenocrysts in the T9-1 basaltic andesite from the SOT (Tables S2–S6, Figure 6). The average depths of ascending magmas for forming the clinopyroxene phenocrysts in the R2 basalt, orthopyroxene phenocrysts in the T2 andesite, orthopyroxene phenocrysts in T6-1 rhyolites from the MOT, and orthopyroxene phenocrysts in the T9-1 basaltic andesite from the SOT were estimated to be 11.3 km (n = 11), 10.9 km (n = 14), 12.7 km (n =28), and 12.6 km (n = 13), respectively. These results suggest that the ascending magmas for forming the R2 basalt, T2 andesite, and T6-1 rhyolites in the MOT and the T9-1 basaltic andesite in the SOT originated from near the crust-mantle transition zone (13–14 km) [62].



Figure 6. Crystallization temperatures and magma source depths for the MOT and SOT volcanic lavas.

The LILE, Pb, and Sr enrichments and the Nb, Ta, and Ti depletions in the MOT and SOT lavas (Figure 4) produced trace element distribution patterns similar to those of the continental crust [114]. If considerable crustal contamination or magma mixing occurred, the ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd of the volcanic lavas would then increase and decrease, respectively, with increasing SiO₂ [12,89]. However, except for the T6-1 and T6-2 rhyolites, the ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd of the MOT and SOT volcanic lavas remained nearly constant with increasing SiO₂ content (Figure 7), which is unlikely to suggest mixing or assimilation

between variably evolved OT magmas, indicating that crustal contamination had a minimal effect on the evolution of MOT and SOT magmas. However, the ⁸⁷Sr/⁸⁶Sr ratios of MOT volcanic lavas could be strongly affected by the involvement of AOC or sediments, resulting in elevated ⁸⁷Sr/⁸⁶Sr ratios in the R2 and T2 volcanic lavas. Furthermore, as observed in the Harker diagrams (Figure S2), the significant negative correlations among SiO₂ and Al₂O₃, CaO, FeO_t, and MgO highlight the fractional crystallization of olivine, clinopyroxene, and plagioclase [115]. P₂O₅ and TiO₂ initially increased and then decreased with increasing SiO₂ content, suggesting that apatite fractionally crystallized after the SiO₂ content reached >60%. Magnetite fractionally crystallized when the SiO₂ content was >55% (Figure S2) [115,116].



Figure 7. (a) SiO₂ vs. ⁸⁷Sr/⁸⁶Sr and (b) SiO₂ vs. ¹⁴³Nd/¹⁴⁴Nd plots showing that crustal assimilation slightly influenced the magmatic evolution of the MOT and SOT volcanic lavas. Data for published mafic samples are from [1,36,50,74]. Data for published felsic samples are from [1,73,74]. Data for samples T6-1, T2, and T6-2 are from this study.

The sample locations, trace element patterns, and Sr-Nd isotopic compositions of the T5-2 trachyandesite (27°32.86' N, 126°59.36' E, 1283 m) and T2 and esite in the MOT (27°32.76' N, 126°58.52' E, 1240 m; 87Sr/86Sr = 0.704522, 143Nd/144Nd = 0.512865) [74] were similar to those previously reported for MOT andesite (287-2: 27°29.50' N, 126°50.00' E, 1380 m; ⁸⁷Sr/⁸⁶Sr = 0.704252, ¹⁴³Nd/¹⁴⁴Nd = 0.512806) [73] (Figure 4c), indicating that samples T5-2, T2, and 287-2 may be derived from a similar magma source [117]. Furthermore, Li, et al. [74] and Shinjo and Kato [73] suggested that the 287-2 and T2 samples were formed by fractional crystallization of basaltic magmas, implying that the T5-2 sample was also produced in this way. Similarly, the sampling location (Table S1), trace element distribution pattern, and Sr-Nd isotope compositions of the R2 basalt from the MOT (27°32.47' N, 126°58.62' E, 1309.7 m; 87Sr/86Sr = 0.704188, 143Nd/144Nd = 0.512763) [74] were similar to those of Type 1 basalt (A6: 27°31.33' N, 126°56.60' E, 967 m; 87Sr/86Sr = 0.704044, 143Nd/144Nd = 0.512827) (Figure 4b) previously reported by [50], indicating that the R2 sample and Type 1 sample A6 from [50] were both produced by the crystallization of a similar magma source and did not suffer notable crustal contamination or undergo any assimilation and fractional crystallization (AFC) processes [50].

However, the T9-1 basaltic andesite from the SOT had lower trace element and REE concentrations than the R2 basalt from the MOT (Table S8, Figure 4a,b), implying that the T9-1 basaltic andesite and the R2 basalt originated from different magmatic sources and evolved through different processes [50]. Furthermore, the trace element distribution pattern of the T9-1 basaltic andesite from the SOT differed from those of other SOT samples

previously reported by [50], suggesting that the source of the SOT magma was heterogeneous and that fractional crystallization of a primitive magma was insufficient to form the different compositions of the SOT magmas [50].

In addition, the T6-1 and T6-2 samples were obtained from the western slope of the trough (Figure 1), and they exhibited the lowest and highest ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd, respectively, among all the OT samples (Figure 7). These pumices all had lower crystallization temperatures and magma depths than the T2 samples obtained from the Iheya Ridge (Figure 6). Zhang, et al. [118] analyzed the geochemistry of the T6-1 and T6-2 pumices and concluded that they did not originate from MOT basaltic rocks through either partial melting or fractional crystallization processes; rather, they were generated from a potassium-rich magma source in the OT. The trace element and REE patterns of the T6-1 and T6-2 rhyolites from the MOT resemble those of Type 2 rhyolites previously reported by [73] (Table S8 and Figure 4e), which can be explained by AFC processes in the MOT basaltic magma [73]. Therefore, the T6-1 and T6-2 rhyolites from the MOT originated from different magma sources and underwent magmatic evolution histories different from those of other MOT samples.

5.2. Influence of Subduction Components Inferred from Trace Elements

The MOT basalts and SOT basaltic andesite were notably enriched in LILEs and Pb and were depleted in HFSEs (Figure 4 and Table S8), indicating that the magma from which these lavas formed had been affected by subduction component input [9]. On the Ba/La vs. Th/Rb and Ce/Pb vs. Ba/La diagrams (Figure 8), all the MOT and SOT whole-rock samples plot are within the subducted sediment zone, indicating that all these volcanic lavas have been affected by subducted sediments. The Ba/La vs. Th/Yb and Ba/Th vs. Th/Nb diagrams (Figure 8) also show that the MOT and SOT trace element ratios trend toward sediment assimilation. All these diagrams suggest that the OT volcanic rocks have been affected by subducted sediment. However, whether slab-derived fluids influenced the mantle beneath the back-arc is still uncertain.



Figure 8. Trace element ratio plots: (**a**) Th/Rb vs. Ba/La, (**b**) Ce/Pb vs. Ba/La, (**c**) Th/Yb vs. Ba/La, and (**d**) Th/Nb vs. Ba/Th for the MOT and SOT volcanic lavas. Compositions of the EPR MORB data are from PetDB database (www.earthchem.org/petdb). Subducted sediment compositions are from [10]. Canary ocean island basalt (OIB) contents are from [119]. Calc-alkaline basalt contents are from [120,121], and the values for the lower crust in the North China Craton are from [1,73,74]. Data for published mafic samples are from [1,36,50,74]. Data for published felsic samples from [1,73,74]. Data for samples R2, T5-2, T6-1, T2, and T6-2 in the MOT and T9-1 in the SOT are from this study.

The Pb content of crustal sediments (19.9 ppm) [10] is two orders of magnitude greater than that of the mantle (0.6 ppm) [12,90], and the addition of dehydration fluids from the subducting plate can increase the contents of fluid-active elements [108]. However, the ratios of fluid-inactive elements to active elements, such as the Nd/Pb ratio (1.33) of the T9-1 basaltic andesite from the SOT, were notably lower than those of MOT samples (basalt = 4.65, andesite = 3.80, trachyandesite = 3.79, and rhyolites = 2.15–3.16), and the Nd/Pb ratios of all MOT and SOT samples were lower than that of the primitive mantle (~7.3) [90], suggesting that a Pb-rich component (i.e., subducted sediment) was present in the OT magma source [12].

The Ba/La vs. Th/Yb and Ba/Th vs. Th/Nb diagrams (Figure 8) show that the MOT and SOT samples trend toward sediment assimilation, which suggests that the OT volcanic lavas were affected mainly by subducted sediment-derived melts and that subduction fluids had less influence on the magma. Furthermore, the Ba/La ratio of the SOT basaltic andesite (20.32) was considerably higher than those of MOT basalt (13.05), andesite (13.79), trachyandesite (14.35), and rhyolites (11.76–14.81). The Ce/Pb ratio of the T9-1 basaltic andesite (2.11) from the SOT was significantly lower than those of MOT samples (4.99–7.05). Compared to the Ba/La ratios of MOT volcanic rocks (11.76–14.81), the Ba/La

ratios (20.32) of T9-1 basaltic andesite from the SOT were closer to the Ba/La ratios of sediment in the Philippines Sea (25.11) [10] (Figure 8), all of which indicate that the SOT magma from which the T9-1 basaltic andesite formed was affected by subducted sediments [8,12–14,74,115].

5.3. Subduction Input of Low-δ⁷Li Components to the OT Magmas

The Li isotope characteristics of back-arc volcanic lavas are known to be potentially influenced by subducted slab-derived components [1,123,124], which include AOC-derived hydrous fluids, subducted sediments, and oceanic crust- and/or sediment-derived silicate melts [125,126]. Although the low δ^7 Li values are not consistent with subduction fluids or the asthenosphere (i.e., MORBs), they may be related to the melting of a dehydrated slab [4,96,124,127]. Previous research has demonstrated that low δ^7 Li values can be interpreted as dehydration signatures resulting from the hydrothermal alteration of oceanic crust (-10.90% to +20.80%) [26,94]. During high-temperature plate subduction processes, low- δ^{7} Li components are released and interact with the upper mantle, thereby resulting in low δ^{7} Li values in the range of -6% to +10% [35]. Other studies have shown that alteration of the upper oceanic crust at high-temperature can also result in low $\delta^7 Li$ values (e.g., -1.7%) [26,128], and forearc serpentinites exhibit δ^7 Li values of less than -6%[35]. Additionally, eclogites, which represent dehydrated oceanic crust, have low $\delta^7 Li$ values (-11‰ to +0.3‰), and the direct melting of such siliceous eclogites could result in low- δ^7 Li melts [28]. Some sediments, especially young oceanic sediments that have not been altered by seawater, also exhibit low δ ⁷Li values (-4.31‰ to +23.33‰) [27]. Thus, fluids with relatively low 87Li values may also be released from subducted sediments [3].

The δ^7 Li values of glass and whole-rock samples from the T5-2 trachyandesite, T2 andesite, R2 basalt in the MOT and the T9-1 basaltic andesite in the SOT were significantly lower than those of whole-rock data for T6-1 and T6-2 rhyolites in the MOT (Table S9). This contrast implies that the magmas from which the T5-2 trachyandesite, T2 andesite, and R2 basalt in the MOT and the T9-1 basaltic andesite in the SOT formed could have been affected by a low- δ^7 Li component that may be released from oceanic sediments or subducted AOC.

Furthermore, previous research has shown that Li isotopes are not fractionated in closed systems at temperatures greater than 350 °C [33,129,130]. Thus, Li isotopes are not expected to have fractionated during OT magma evolution [33,131,132]. However, the δ⁷Li values of clinopyroxene in the T5-2 trachyandesite (-0.83%) from the MOT and T9-1 basaltic andesite (0.49%) from the SOT and of plagioclase in the R2-1 basalt (-0.71%) from the MOT and T9-1 basaltic andesite (-5.05%) from the SOT were lower than those of olivine, orthopyroxene, glass (Table S9), and MORBs (+1.50% to +6.85%) [22,33,91–93] (Figure 5a,b). This difference suggests that the Li isotope compositions of plagioclase and pyroxene may have been modified by a low-δ⁷Li fluid or melt during magma evolution and that the ⁶Li content was enriched in clinopyroxene and plagioclase during mineral crystallization and mantle metasomatism. Overall, the negative Li isotope fractionation (Δ^7 Lix-ol < 0) between silicate minerals and olivine during mineral crystallization may have been due to the low-δ⁷Li components that are released from oceanic sediments or AOC.

5.4. Subduction of Low-and High-δ¹⁸O Components

For volcanic rocks, magma differentiation from mafic to felsic compositions at high temperatures does not result in significant oxygen isotope fractionation (usually less than 0.3–0.4‰) (e.g., from 5.8‰ for basalts to 6.1‰ for rhyolites at ~90% differentiation) [133,134]. The δ^{18} O values of glass (6.31‰) and plagioclase (5.99‰) in the T5-2 trachyandesite, glass (6.21‰) and orthopyroxene (5.86‰) in the T2 andesite, the whole-rock samples of T5-2 (6.69‰) trachyandesite, T6-1 (5.84‰), and T6-2 (6.08‰) rhyolites from the MOT (Table S9) were all higher than those of MORBs (+5.30 to +5.80‰) [110,111] (Figure 5c); this feature also indicates that these volcanic lava samples were produced by high- δ^{18} O magmas [135,136]. The high δ^{18} O values of the evolved magmas may be related to

crustal contamination; however, no correlations were observed between the Sr-Nd isotopes and SiO₂ concentrations in OT lavas (Figure 7), suggesting that little crustal contamination occurred [11,36,50,73,74]. Alternatively, the high- δ^{18} O signatures could be due to the subduction of δ^{18} O-rich components [110,137]. Marine carbonates (δ^{18} O = 25–32‰), siliceous oozes (δ^{18} O = 35–42‰), and pelagic clays (δ^{18} O = 15–25‰), which are major components of sediments subducting beneath the volcanic arc, all have higher δ^{18} O values than MORBs [2]. Therefore, the addition of subducted sediments to the magma could explain the higher δ^{18} O values of MOT volcanic rock samples.

However, O isotopes can be used to constrain the contribution of slab melts to island arc magmas, thereby indicating the origin of a melt [2]. The δ^{18} O values of the R2-1 (5.41– 5.79%) and R2-2 (5.38-5.78%) basalts from the MOT fell within the MORB range (5.3-5.8%), implying that the volcanic lava samples in the MOT originated from mantle-derived magma (Figure 5c) [111,138]. The whole-rock and mineral δ^{18} O values (5.15–6.31‰) of the T5-2 trachyandesite varied significantly (Figure 5c) and may have been affected by different magmatic components [2] during magma ascent. Moreover, the δ^{18} O values of whole-rock samples (5.29‰), olivine (4.83‰) and clinopyroxene (4.99‰) from the T9-1 basaltic andesite in the SOT were lower than those ([5.30–5.80‰) [110,111] of MORBs (Figure 5c). This result indicates the presence of a low- δ^{18} O component during magma evolution [37,139,140]. Furthermore, AOC had a large range of δ^{18} O values (+2‰ to +14‰) [141], and the low δ^{18} O values of ocean island basalts (OIBs) and MORBs can be explained by the addition of subducted oceanic crust to the magma source region [142–145]. Therefore, the low δ^{18} O values of the T9-1 basaltic and esite from the SOT are attributed to siliceous melts or fluids derived from subducted AOC [143,145]. Furthermore, the δ^{18} O value (5.50%) of orthopyroxene in the T9-1 basaltic andesite from the SOT was lower than that (5.86‰) of orthopyroxene in the T2 and esite from the MOT, which was consistently related to the varying crystallization temperatures (Tables S2 and S3), suggesting that the high-temperature (1095–1138 °C) orthopyroxenes were characterized by ¹⁶O enrichment in the SOT, whereas the low-temperature (1020–1095 °C) orthopyroxenes were characterized by 16O depletion in the MOT.

As with the Li isotopes, the δ^{18} O values of plagioclase and glass were higher than those of MOT and SOT olivine, clinopyroxene, and orthopyroxene (Table S9 and Figure 5c), and all the samples exhibited positive O isotopic fractionations between other minerals and olivine ($\Delta^{18}O_{x-0} > 0$). This feature suggests that compared to olivine, plagioclase and pyroxene preferentially incorporated ¹⁸O rather than ¹⁶O during silicate mineral crystallization, which indicates that the O isotope compositions of plagioclase and pyroxene may have been modified by a high- δ^{18} O fluid or melt [137].

5.5. Contribution of High- δ^{26} Mg Slab Fluids

The δ^{26} Mg values of MOT volcanic samples (-0.21‰ to -0.28‰) fell within the range of MORBs (-0.26‰ ± 0.07‰) [107] (Figure 5e). The δ^{26} Mg values of the T2 andesite (-0.20 to -0.12‰) were similar to those of the T5-2 trachyandesite (-0.23‰ to -0.12‰) (Figure 5e), both of which were slightly higher than those of MORBs (Figure 5e). The δ^{26} Mg values (-0.16‰ to -0.09‰) of the T9-1 basaltic andesite in the SOT, which were slightly higher than those in MOT volcanic lavas (Table S9), were also higher than those of MORBs (Figure 5e), all of which indicates that these lavas may have originated from high- δ^{26} Mg magmatic components [5,39,146].

Previous studies have shown that higher Mg isotopes in subduction zones may originate from seawater alteration, chemical weathering, dissolution of continental crust, partial melting of oceanic crust, and/or sediment and slab dehydration [107,108,147–151]. Dissolution of continental crust can be ruled out due to the absence of correlations between the Sr-Nd isotopic compositions and the indices of magma differentiation (e.g., SiO₂; Figure 7). Furthermore, volcanic lavas can have isotopically heavier δ^{26} Mg values than MORBs if they are generated by the melting of subducted oceanic crust containing a garnet residual phase because garnet has much lower δ^{26} Mg values [41,42,108,152]. However, because the chemical compositions of our samples were quite different from those of slab products (e.g., adakites) (Figure 4), the possibility of slab melting can also be excluded. Therefore, either subducted sediments or slab dehydration may explain the high δ^{26} Mg values of volcanic lava samples analyzed in this study.

On average, forearc sediments have heavy Mg isotope compositions (-0.10 ± 0.61) that could provide a source for volcanic lavas with heavy Mg isotope compositions [108]. Moreover, the δ^{26} Mg values of the Avacha volcanic lavas from the Kamchatka Peninsula range from -0.25% to -0.06% (average = $-0.18\% \pm 0.10\%$ (2SD)) [153], which has been interpreted as the result of the lower mantle being affected by subduction fluids released by the Pacific slab [148]. Similarly, the δ^{26} Mg values of the Avacha volcanic lavas are consistent with those of whole-rock samples from MOT andesite (-0.17% to -0.12%), trachyandesite (-0.16% to -0.12%), and basalt (-0.28% to -0.20%) and SOT basaltic andesite (-0.11% to -0.09%), implying that fluids released from subducted sediment or oceanic crust contributed to the OT magma source.

5.6. Mixing of Subduction Components

The above discussion suggests that the OT magma source was influenced by both subducted sediments and AOC [1,47,50]. A residual slab endmember, represented by eclogite, and an AOC endmember interacting in different proportions with a MORB-producing magma could produce lavas with different δ^7 Li, δ^{18} O, and δ^{26} Mg values [7,20,33]. According to the δ^{18} O vs. δ^7 Li and δ^{26} Mg vs. δ^7 Li diagrams (Figure 9), OT volcanic lavas with different δ^7 Li, δ^{18} O, and δ^{26} Mg values can be produced by interactions between MORB and different proportions of subducted sediment (i.e., low δ^7 Li and high δ^{18} O and δ^{26} Mg) and AOC (i.e., high δ^7 Li and low δ^{18} O), wherein the AOC, oceanic sediments, and mantle wedge (i.e., MORB) are the endmembers contributing to the MOT and SOT magmas. The whole-rock and glass data from the R2 basalt in the MOT and the T9-1 basaltic andesite occur in the SOT plot between subducted sediments and AOC, indicating that the contributions from AOC and sediments were 80% to 92% and 20% to 8%, respectively; therefore, the mixing of different subduction components can produce Li, O, and Mg isotope characteristics in the R2 basalt from the MOT and the T9-1 basaltic andesite from the SOT (Figure 9 and Table S9).



Figure 9. Plots of (a) δ^{18} O vs. δ^{7} Li and (b) δ^{26} Mg vs. δ^{7} Li for the MOT and SOT samples. Isotopic compositions (δ^{18} O = 2‰, δ^{7} Li = 11‰, and δ^{26} Mg = -0.25‰) of AOC are from [33,39,137], respectively. Isotopic compositions (δ^{18} O = 22‰, δ^{7} Li = -2.1‰, and δ^{26} Mg = -0.1‰) of subducted sediments are from [7,33,108], respectively. Isotope compositions (δ^{18} O = 5.5‰, δ^{7} Li = 6.5‰, and δ^{26} Mg = -0.35‰) of mid-ocean ridge basalts (MORBs) are from [22,107,110], respectively.

5.7. Implications of Differences in Plate Subduction

As discussed above, the SOT magma from which the T9-1 basaltic andesite formed was influenced by more slab-derived fluids, although the supporting data are limited. However, the thermal structure of a subduction zone is the key to determining the depth at which the subducted slab dehydrates [154–157], and this thermal structure is mainly determined by the depth of the subducted slab, age of the subducting plate, convergence rate, subduction zone geometry (especially the subduction angle), subduction zone shear heating rate, and nature of the mantle wedge [11,65,123,155,157]. Furthermore, dehydration usually decreases with increasing subduction depth [47,123,158], and most subducted slabs dehydrate considerably at a depth of ~80 km, leaving <1% of the water to be carried deeper by the subducting slab [154]. The subducting plates lose notable volumes of water at relatively shallow depth (modeled here at a depth of 80 km), and the further dehydration of most slabs is only minor (e.g., Kamchatka and Calabria) (Figure S4).

The OT is influenced by the subduction of the PSP, and the difference between the subduction rates of the SOT and the MOT is relatively small [11,50,56,69]. Therefore, the difference between the subduction rates likely had only a limited impact on plate dehydration [47,50,157]. Furthermore, the subduction direction of the PSP is nearly perpendicular to the axis of the MOT and becomes oblique to the SOT [11,50,65]. Although the slab subduction angle beneath the SOT is higher compared to that beneath the MOT, this small difference does not significantly influence slab material transport [11,47,50]. Moreover, the difference between the crustal thicknesses of the MOT and the SOT (i.e., ~16 and ~14 km, respectively) is negligible when compared to the subduction depths of the PSP in the MOT and SOT (~200 and ~150 km, respectively), suggesting that the slight difference between the crustal thicknesses of the MOT is not enough to cause different degrees of slab dehydration. However, this difference may reflect the degree of crustal contamination when the magma ascends from a deep magma chamber.

The Ryukyu subduction zone has a fairly rapid subduction rate (~82 mm/a) [159], which is in line with that of cold subduction zone structures [47], suggesting that large degrees of dehydration occurred when the slab reached depths of ~80 to 100 km, at which point sediment fluid/melt entered the mantle wedge [157]. Moreover, the ascending magma from which the T9-1 basaltic andesite in the SOT formed corresponds to a Wadati-Benioff zone depth of ~150 km, and the MOT has Wadati-Benioff zone depths of ~150 to 200 km (Figure 1). However, compared with the PSP subducting slab (depths: ~150 to 200 km) in the MOT, the PSP subducting slab in the SOT exhibits a significantly shallower subduction depth (~150 km), which is consistent with the variation in the subduction angle, i.e., nearly perpendicular beneath the MOT and becoming progressively more oblique beneath the SOT [11,50,65]. This variation indicates that the subducting slab (depth of ~150 km) beneath the SOT was closer to the ascending magma (avg. depth = 12.6 km) that formed the T9-1 basaltic andesite in the SOT than the subducting slab (depth of ~200 km) beneath the MOT. The deep ascending magmas (depth: 8.4–16.1 km and ~8.0–16.9 km) that formed the clinopyroxene and orthopyroxene phenocrysts with high crystallization temperatures (1095–1138 °C and 1111–1137 °C, respectively) in the mafic volcanic lavas (SOT basaltic andesite T9-1 and MOT basalt R2) were closer to the subducted slab than the shallow ascending magma (depth: 3.7–7.1 km) that formed the orthopyroxene phenocrysts with low crystallization temperatures (844-858 °C) in the felsic volcanic lavas (MOT rhyolites T6-2). These data imply that the deeper the ascending magma is, the closer it is to the subducted slab, and the stronger the influence of the subducted components on the ascending magma (Figure 10).





Figure 10. Schematic diagram showing the different contributions of subduction components to the (a) MOT and (b) SOT.

6. Conclusions

(1) The T9-1 basaltic andesite in the SOT had lower trace element contents than the MOT samples and exhibited stronger Nb, Ta, and Ti depletions and a positive Pb anomaly. The Nd/Pb and Ce/Pb ratios of the SOT basaltic andesite were significantly lower than those of the MOT volcanic lavas, while the Ba/La ratio of the SOT basaltic andesite was significantly higher, indicating that the magma from which the T9-1 basaltic andesite from the SOT formed was influenced by subduction components and experienced an injection of sediment components.

(2) The δ^7 Li values of the whole-rock samples from the T6-1 and T6-2 rhyolites in the MOT were higher than those of whole-rock and glass separates from the T5-2 trachyandesite, T2 andesite, and R2 basalt in the MOT and the T9-1 basaltic andesite in the SOT. These results imply that the magmas that formed the R2 basalt in the MOT and the T9-1 basaltic andesite in the SOT could have been affected by a low- δ^7 Li component that may have been released from oceanic sediments or subducted AOC. The δ^7 Li values of plagio-clase and clinopyroxene phenocrysts were lower than those of glass and olivine in the MOT and SOT volcanic lavas, suggesting that ⁶Li was preferentially removed from the magma and incorporated into plagioclase and clinopyroxene, resulting in relative enrichment in heavy Li isotopes in olivine and glass.

(3) The SOT and MOT magmas were influenced by low- δ^{18} O AOC fluids or melts and high- δ^{18} O sediment components, respectively. The δ^{18} O values of plagioclase and glass were higher than those of olivine, clinopyroxene, and orthopyroxene phenocrysts in the MOT and SOT volcanic lavas, indicating that ¹⁶O was preferentially removed from the magma and incorporated into olivine, clinopyroxene, and orthopyroxene, resulting in the relative enrichment in heavy ¹⁸O isotopes in plagioclase phenocrysts and glass.

(4) The δ^{26} Mg values of clinopyroxene, orthopyroxene, and glass were higher in the T9-1 basaltic andesite from the SOT than those in the MOT volcanic lavas, and the δ^{18} O values of the T9-1 basaltic andesite from the SOT were lower than the δ^{18} O of whole-rock volcanic samples from the MOT. These differences imply that the influence of subduction components with high δ^{26} Mg and low δ^{18} O values was stronger in the SOT than in the MOT, which may have occurred because the amount of subducted sediment and AOC dehydration fluids injected into the magma in the SOT was larger than that in the MOT.

(5) The δ^{18} O and δ^{26} Mg values of pyroxenes in the SOT were lower and higher than those in the MOT, respectively, which is consistent with the variations from the SOT to the MOT in the crystallization temperatures, pressures, and depths of the ascending mag-

mas from which volcanic lavas formed. These results suggest that high-temperature pyroxenes originating from a deep magma (avg. depth = 12.6 km, n = 13), which was located near the crust-mantle transition zone (13–14 km), are characterized by ¹⁶O and ²⁶Mg enrichments in the SOT. Low-temperature pyroxenes originating from a shallow magma (avg. depth = 10.9 km, n = 14) are characterized by ¹⁶O and ²⁶Mg depletions in the MOT. However, the distance between the subducting slab and the overlying magma may have played a significant part in controlling the differences between the MOT and SOT in the amounts of subduction components injected into the magma. The deeper the magma is, the closer it is to the subducted slab, and the stronger the effects of the subducted inputs on the magma.

Supplementary Materials: The following are available online at www.mdpi.com/article/10.3390/jmse10010040/s1, Figures S1–S4, Tables S1–S9, and Text S1. These materials include (1) additional analytical methods, (2) major elements, trace elements, and isotopes data obtained in this study, and (3) additional plots for major elements and isotopes.

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