







Older magma at Aso caldera than at Unzen stratovolcano in south west Japan as recorded through helium isotopes

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Stratovolcanoes form conical mountains by ejecta and lava, while caldera volcanoes have depressions caused by post caldera collapse. Few studies have investigated relationships between this morphology difference and discharged fluid geochemistry. Here we present helium isotope ratios of geothermal fluids and groundwater inside and outside the caldera of Mt. Aso, Japan, and compare these to helium ratios in groundwater at Mt. Unzen, a stratovolcano in the same tectonic field. We find that helium isotopes decrease in proportion to distance from the central part of the volcano. However, the helium isotopes of inferred magmas are lower in Mt. Aso relative to Mt. Unzen. We combine this result with seismic tomography data and suggest that Mt. Aso may not be receiving new mantle material to the magma reservoir. Thus, we conclude that the caldera volcano Mt Aso probably features older magma and more developed geothermal systems than the stratovolcano Mt. Unzen.

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Mt Aso is a caldera volcano located in Kyushu Island, southwest Japan, where caldera volcanoes have a large circular depression in or near the center of the mountain body, usually surrounded by a steep wall. Mt Unzen is a composite stratovolcano in the same region, where stratovolcanoes are formed by repeated outpouring of lava and ejection of volcanic debris, which alternately accumulate to form large conical edifices. Topographically, craters of stratovolcanoes are similar to caldera volcanoes, but have a much smaller diameter¹. These volcanoes formed in subduction zones are different in shape and scale from shield volcanoes created in hotspot areas such as Hawaii and Iceland². Comparing these morphological differences, the average volume of ejecta from Japanese caldera volcanoes is larger than that of stratovolcanoes³. Because the chemical composition of ejecta varies with the evolving stage of each volcano, a clear distinction between caldera and stratovolcanoes is difficult to make. Studies of Crater Lake volcano⁴ and Hakone volcano⁵ have shown that a stratovolcano forms first, but later a large eruption releases a large amount of magma, and the magma chamber becomes a void, into which the central part of the volcano sinks, forming a huge caldera with a diameter of over 10 km. There are a couple of questions such as the age of the magma chamber in stratovolcanoes and caldera volcanoes, the structural differences in the surrounding hydrothermal systems, and whether stratovolcanoes eventually evolve into caldera volcanoes.

Geochemical studies of volcanic-hydrothermal systems are often based on stable isotopes of light elements such as carbon, oxygen, and hydrogen, which are volatile elements that migrate with geothermal system⁶. Among these, the helium isotope ratio is the most sensitive parameter to magmatic activity. This is because helium is a noble gas and does not change chemically, helium-3 is primordial derived from the mantle in the Earth's deep interior, and helium-4 is radiogenic derived from the decay of U and Th in crustal materials⁷. Helium isotope ratios are known to vary widely in subduction zones and are closely related to the geotectonics of the sampling sites⁸. Around independent stratovolcanoes, helium isotope ratios generally change from mantle values to crustal values with increasing distance from the central cone⁹. In the vicinity of caldera volcanoes, on the other hand, data on the geographic distribution of helium isotope ratios are very scarce. In this study, we focus on Aso volcano, a representative caldera volcano in Japan, to clarify the distribution of helium isotope ratios inside and outside the caldera. The obtained data are compared with those of Unzen volcano, a stratovolcano located in the same tectonic field, to discuss the age of magma and the degree of hydrothermal system development. This study may provide crucial information of differences between system of caldera volcano and stratovolcano.

Results

Aso volcanic activity began about 300,000 years ago. A total of four major eruptions occurred about 70,000 to 80,000 years before the current Aso caldera was formed. They are called Aso-1, Aso-2, Aso-3, and Aso-4. The present caldera shape was completed after the eruption of Aso-4. The Aso caldera was formed immediately after the eruption of Aso-1 and gradually enlarged during the subsequent eruptions. After the pyroclastic flow of Aso-4, the next volcanic activity started with a slight time lag and successively formed the central cluster of volcanoes. Among these, Nakadake is still active today with intermittent eruptions. The post-caldera activity involved a wide range of magmas from basalts to rhyolites, resulting in changes in the shape and structure of the volcano (ref. ¹⁰). We collected a total of 28 hot spring water,

groundwater, and hot spring gas samples at 9 sites within the caldera and 9 sites outside the caldera in the Kumamoto Plain, the Kyushu Island, southwest Japan. All measured helium isotope ratios and helium/neon ratios are shown in Table S1, along with sampling dates, water temperatures, and well depths. $^4\text{He}/^{20}\text{Ne}$ ratios range from 0.40 to 27.6, $^3\text{He}/^4\text{He}$ ratios are 0.62 Ra (where Ra is the helium isotope ratio of the atmosphere and has a value of 1.382×10^{-6} , (ref. ¹¹)) to 7.00 Ra. These data can be explained by a mixing of three components: mantle-derived helium, crustal helium, and atmospheric helium. These are typical subduction zone helium isotope values and there is nothing unusual about them⁸. The $^3\text{He}/^4\text{He}$ ratio, corrected for the atmospheric component using the $^4\text{He}/^{20}\text{Ne}$ ratio, varied from 0.57 to 7.17 Ra. There is no clear positive correlation between water temperature and $^3\text{He}/^4\text{He}$ ratio. Therefore, the heat source of hot springs in this region cannot simply be attributed to magma. There may also be a heating effect due to the geothermal gradient in the crust.

Table S2 shows the chemical composition of free gas from hot springs and gaseous phase extracted from hot spring water and groundwater. In addition, Table S2 indicates the $^{40}\text{Ar}/^{36}\text{Ar}$ ratios, $\delta^{15}\text{N}$ values, and $\delta^{13}\text{C}$ values for gases with sufficient amounts of argon, nitrogen, and carbon dioxide for which isotopic ratios could be measured. Nitrogen and carbon dioxide were the main gaseous components, and more than 10% methane was found in two samples. $^{40}\text{Ar}/^{36}\text{Ar}$ ratios were close to the atmospheric value of 295.5 in many samples, and even when the largest isotope ratios were measured with 10% excess ^{40}Ar . Nitrogen isotope ratios are mostly 0‰ with negative anomalies in some samples, but appear to be essentially atmospheric in origin. Carbon isotope ratios varied from -26.3‰ to -3.9‰ . These values could be explained by a mixture of oxidized organic matter in sedimentary layers, marine carbonates, and mantle-derived carbon¹² together with depletion by calcite precipitation.

Unzen is the collective name for a group of volcanoes that extend 20 km from east to west and 25 km from north to south, centering on Fugendake. Unzen volcanoes are mainly composed of two volcanic groups: older volcanoes such as Takadake and Kusenbu-dake, and younger volcanoes such as Fugendake, Mayuyama, Myokendake, and Nodake. K-Ar ages range from 500 to 100 ka for the older ones and after 100 ka for the younger ones. There is a time difference of about 10,000 years between the two. The total volume of ejecta converted to lava is about 44 km^3 , of which the volume of the older ejecta is 36 km^3 , overwhelmingly larger than that of the younger ejecta (8 km^3). This means that the major part of Unzen was completed in the older volcanic stage. The younger volcanoes are, in order, Nodake, Myokendake, Fugendake, and Mayuyama. Ejecta from each volcano are classified as lava and pyroclastic flow deposits. Fugendake began its latest eruption in November 1990. (ref. ¹³). After the first round of review on this work, we conducted a field work in the Unzen volcanic geothermal region in August 2022 and collected 7 hot spring water and gas samples at 7 sites in Fig. 1. All measured helium isotopes and helium/neon ratios are listed in Table S3 together with data referred from Morikawa et al. (ref. ¹⁴). The corrected $^3\text{He}/^4\text{He}$ ratios varied from 1.0 to 7.3 Ra, generally consistent with those of Aso samples.

Discussion

From a geotectonic perspective, Aso volcano is located in the Beppu-Shimabara graben zone, which is considered to divide Kyushu Island into northern and southern parts. The graben zone is formed by three factors: extension of the Okinawa Trough to the north, extension of the Median Tectonic Line to

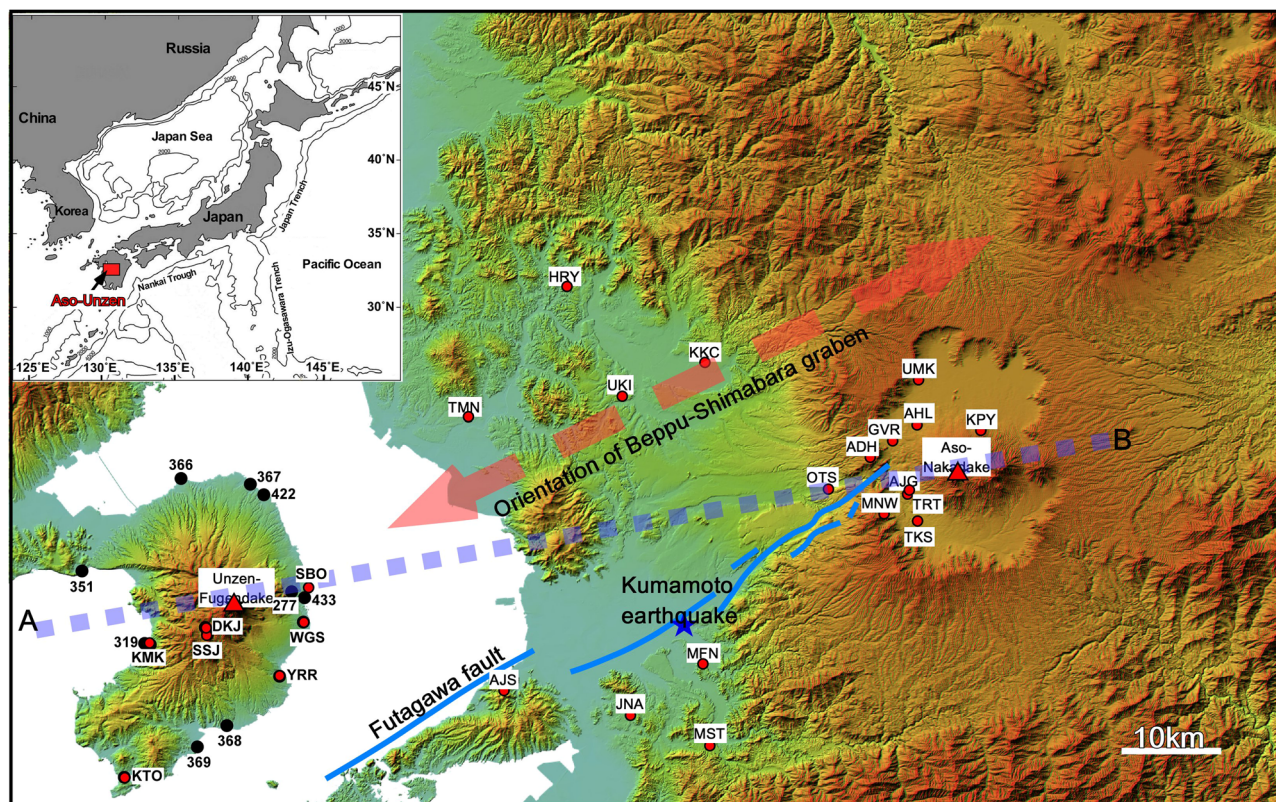


Fig. 1 Location of sampling sites. Location of sampling sites (red dots) at the inside and outside of Mt. Aso caldera together with around Mt. Unzen. Those of black dots are referred from Morikawa et al. (ref. ¹⁴). Abbreviations of symbols are explained in Table S1 and S3. Blue star and thick dashed line show epicenter of the 2016 Kumamoto earthquake (M7.3) and the location of seismic tomography image in Fig. 3, respectively. Orientation of the Beppu-Shimabara graben and Futagawa faults are shown by thick dotted red and blue solid curves. The base map is modified from Digital Japan Portal Web Site, Geospatial Information Authority of Japan (<http://maps.gsi.go.jp>).

the west, and hot mantle upwelling, based on detailed 3-D structure analysis using seismic tomography¹⁵. This graben zone contains several complex stratovolcanoes such as Tsurumidake, Kuju, and Unzen that exhibit Quaternary magmatic activities. Among these volcanoes, Aso is unique because it is a caldera volcano in terms of morphology. Figure 2a shows the relationship between the distance from the cone of Aso/Nakadake to the sampling point in Fig. 1 and the air corrected helium isotope ratios. The $^3\text{He}/^4\text{He}$ ratios tend to decrease with distance from the crater. This result is consistent with the previous distribution of helium isotope ratios around stratovolcanoes⁹. This trend is due to the dilution of mantle-derived helium in the fluid with distance by helium produced by radioactive decay of U and Th in the aquifer rocks. The mean and standard deviation of the inner caldera sample is 5.02 ± 1.16 Ra (1σ), which is higher than the outer value of 1.73 ± 1.05 Ra (1σ). Within the caldera, No. 4-MNW shows uniquely low helium isotope ratios. This sample is free gas in spring water collected at the surface. There is a thick sedimentary layer of up to a hundred meters at this site, which could be explained by the hypothesis that while the fluid was ascending through this sedimentary layer, $^3\text{He}/^4\text{He}$ was lowered by the steady addition of radiogenic helium due to the decay of U and Th in the sedimentary rocks¹⁶.

In general, the geographic variation of helium isotope ratios around stratovolcanoes can be explained by the Hydrodynamic Dispersion (HD) model, which is possibly over-simplified¹⁶. Assuming that the system in which mantle helium flow is from a point source, steady state, and homogeneous in radial direction, the helium isotope ratio at distance (r) from the central cone can

be estimated by the following equation:

$$^3\text{He}/^4\text{He}(r) = (^3\text{Pr} + ^3\text{C}/r)/(^4\text{Pr} + ^4\text{C}/r) \quad (1)$$

where ^3P , ^4P , ^3C , and ^4C are the amount of ^3He and ^4He produced per unit time by nuclear reactions and radioactive decay, and the concentration of ^3He and ^4He in the magma below the central crater, respectively. For a typical sedimentary layer in a subduction zone, we can assume that $^3\text{P} = 3 \times 10^{-2}$ atoms $\text{m}^{-3} \text{sec}^{-1}$ and $^4\text{P} = 1.5 \times 10^6$ atoms $\text{m}^{-3} \text{sec}^{-1}$ (ref. ¹⁶). These values were calculated by $\text{U} = 3$ ppm and $\text{Th} = 10$ ppm, and radiogenic $^3\text{He}/^4\text{He}$ ratio of 2×10^{-8} . In Kumamoto plain located between Mt. Aso and Unzen (Fig. 1), aquifer rocks are composed of biotite gneiss, pelitic, mafic and psammitic schists. Their U and Th concentrations were not measured, but their ^4He abundance of 1.9×10^{-6} $\text{cm}^3\text{STP/g}$ was documented (ref. ¹⁷). It is difficult to estimate U and Th contents from the ^4He abundance, because information of age and retention rate of ^4He in the rock are necessary. In the case of groundwater of Kobe region in southwest Japan, aquifer rock showed $\text{U} = 2.8$ ppm and $\text{Th} = 10.1$ ppm (ref. ¹⁸) in addition to ^4He abundance of 2.1×10^{-6} $\text{cm}^3\text{STP/g}$ (ref. ¹⁹). The ^4He abundance is similar to that of Kumamoto aquifer rock. Thus, the assumption of $\text{U} = 3$ ppm and $\text{Th} = 10$ ppm is somehow justified.

The curve in Eq. (1) can be fitted to the observed values in Table S1 with ^3C and ^4C as unknown parameters. The red dotted line in Fig. 2a is the optimal fitting curve excluding the anomalous No. 4-MNW, and the blue dotted line is the result adding the singularity. The two fitted curves differ within the caldera, but agree outside the caldera at a distance from the central cone. We will tentatively proceed with the best-fit curve excluding the

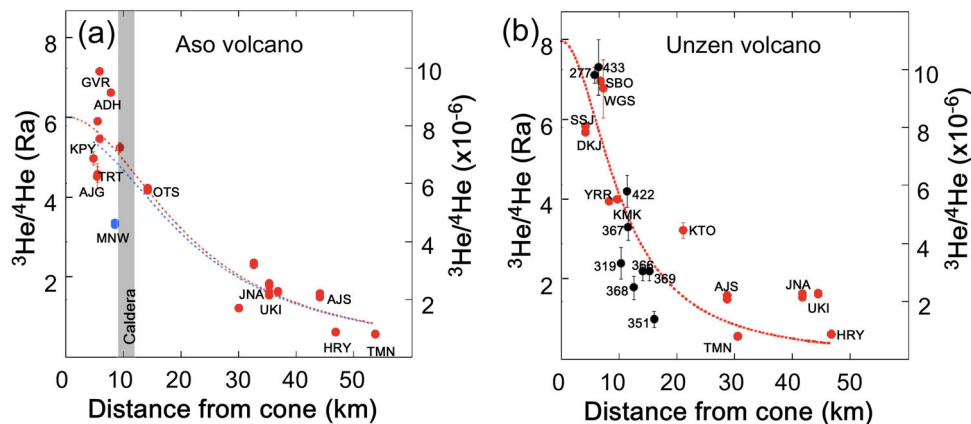


Fig. 2 Relationship between the distance from the volcanic cone to the sampling site and helium isotopes corrected for air contamination. **a** The relationship from Aso volcano where red and blue circles show water and gas data. Blue circles show exceptionally low helium isotopes at No. 4-MNW. Blue and red dotted curves are the best fit to the data estimated by the Hydrodynamic Dispersion (HD) model with No. 4 and without No. 4, respectively. **b** The relationship from Unzen volcano where red and black circles show water and gas data. Black circles data are referred from Morikawa et al. (ref. ¹⁴) together with data obtained in this work (red circles). Red dotted curve shows the best fit to the data by the HD model. The Error assigned to the red circle data obtained in this study is 1 sigma (SE), while the error assigned to the black circles was derived from calculations by Morikawa et al. (ref. ¹⁴).

singularity, which seems to have been influenced by thick sedimentary layers.

By setting $r = 0$ in the curve obtained from the HD model, the helium isotope ratio at the Nakadake cone is estimated to be 6.01 ± 1.51 Ra (1σ error). Due to sampling difficulties, the helium isotope ratios of fumarolic gases at the crater have not been measured, so a direct comparison with this value is not possible. Some samples in the caldera, such as No. 1-ADH and No. 7-GVR, show helium isotope ratios higher than this 6.01 Ra (Fig. 2a). However, these data are within a 1σ error of the estimated maximum. This problem will be resolved when the central cone fumarole is available. The helium flux (emitted per unit time and unit area) can be estimated from the HD model. Due to the nature of Eq. (1), the fluxes of ^3He and ^4He cannot be calculated for $r = 0$. Therefore, in the case of the 2014 eruption of Mt. Ontake, the flux at a distance of 1 km from the point source was used by the dike model directly below the volcano²⁰. Assuming $r = 1$ km in this study as well, the helium-3 flux at Aso volcano can be calculated to be 5.7×10^6 atoms $\text{m}^{-2} \text{sec}^{-1}$. This value is about twice larger than that of Ontake volcano just after the 2015 eruption, which is 2.8×10^6 atoms $\text{m}^{-2} \text{sec}^{-1}$ (ref. ²⁰).

Unzen volcano is a composite stratovolcano located in the same Beppu-Shimabara graben zone as Aso volcano. Therefore, they are in the similar geotectonic field and are considered to be optimal stratovolcanoes to compare with caldera volcanoes. The crater of Unzen/Fugendake is located about 75 km west of the cone of Aso/Nakadake (Fig. 1), and several helium isotope ratios of the surrounding hot springs and groundwater have been measured in this work. We combined our data with those reported by Morikawa et al. (ref. ¹⁴) and adopted by Sano and Fischer⁹. Their sampling points are shown around Unzen volcano in Fig. 1. These helium isotope ratios are plotted in Fig. 2b as a function of distance from the Fugendake crater after the air helium correction. The helium data in the Kumamoto Plain (Nos. 12-UKI, 13-HRY, 14-TMN, 15-AJS, and 16-JNA) measured in this study are also plotted with the calculated distance from the Fugendake cone. Based on the geographical distribution of these data, the HD model was fitted with the same parameters as for the Aso volcano. The dotted red line in Fig. 2b incorporates all the data. Some samples from the Kumamoto Plain, which are more than 25 km away from the Fugendake crater, show a large deviation from the model curve and apparent helium-3 excess. This excess can be considered to be the effect of mantle helium

brought from the Aso volcano. On the other hand, in Fig. 2a, plotted at a distance from the Aso/Nakadake cone, the data for the Kumamoto Plain are consistent with the model curve (dotted red line). In addition, the deviations from the model curve are balanced by positive and negative differences. Therefore, it can be interpreted that the influence of mantle helium brought from Unzen volcano is small in these samples. In other words, the hot springs and groundwater in the Kumamoto Plain are more strongly influenced by the Aso volcano, although the distance from the crater is almost the same.

The apparent lower helium isotope ratios in the Kumamoto Plain samples than those near the Aso and Unzen volcanoes may be related to structural heterogeneities in the crust and upper mantle. Figure 3 shows P- and S-wave velocity (V_p , V_s) images of the crust and upper mantle at 0–100 km depths along the profile A-B as shown in Fig. 1, derived from a 3-D tomographic model of the Kyushu subduction zone¹⁵. In this area, V_p and V_s exhibit strong lateral variations of -6% to $+6\%$. In both V_p and V_s images, a high-velocity anomaly (blue) exists beneath the Kumamoto Plain between the Aso and Unzen volcanoes, which extends to a depth of about 30 km. This high-velocity anomaly may reflect a strong part of the crust and block the fluid passage between the near-surface and the deep subsurface, and thus inhibits the ascent of mantle fluids, which is probably the reason for the low helium isotope ratios in the Kumamoto Plain. Directly to the east of the hypocenter of the 2016 M7.3 Kumamoto earthquake (star), there is a thin low-velocity anomaly (orange) at depths of 0–27 km. The Futagawa fault zone is located directly above this low-velocity anomaly, where higher helium isotope ratios than the surrounding area have been observed^{17,21}. These results indicate that some mantle-derived fluids are probably still being supplied from the upper mantle. Similarly, a prominent low-velocity anomaly exists in the crust beneath Unzen volcano, which is connected with a large low-velocity zone in the upper mantle (Fig. 3). Although the magma chamber under Unzen volcano cannot be identified with this resolution, it is highly possible that magma is still being supplied to Unzen volcano from the upper mantle. On the other hand, a high-velocity anomaly (blue) appears in the upper crust beneath Aso volcano (Fig. 3). This feature is discussed in the followings.

From the HD model curve of helium isotope ratios around Unzen volcano, the helium isotope ratio at the Fugendake crater can be calculated to be 7.95 Ra by setting $r = 0$. This is larger

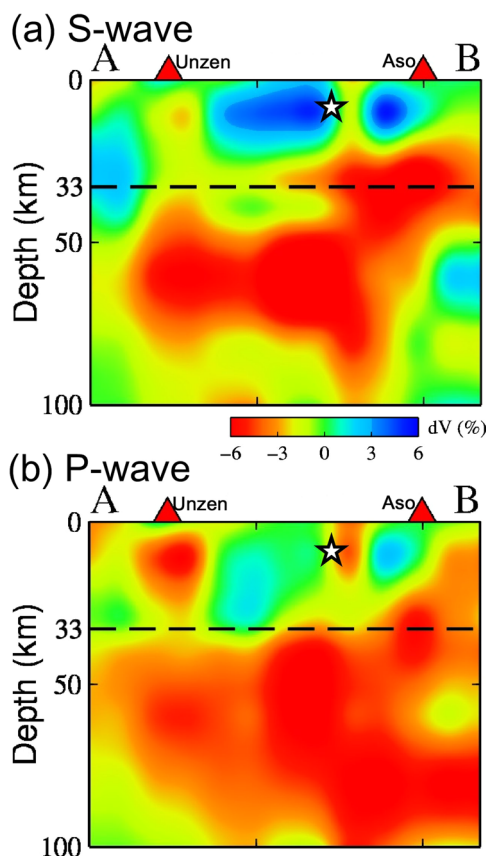


Fig. 3 Vertical cross-section of seismic tomography along the profile A-B in Fig. 1. The colors show perturbations of **a** S-wave velocity (V_s) and **b** P-wave velocity (V_p) relative to the average velocity at each depth. The scale of velocity perturbations (in %) is shown below **(a)**. The star symbol denotes hypocenter of the 2016 Kumamoto earthquake (M7.3). Red triangles denote locations of Unzen and Aso volcanoes. The black dashed line denotes the Moho discontinuity.

than the estimated value of 6.0 Ra for $r = 0$ at Aso Volcano. If 8 Ra, the helium isotopic ratio in quenched glass of mid-ocean ridge basalt, is representative of the upper mantle²², and the radiogenic ratio of ~ 0.02 Ra is representative of the crust²³, the helium in Unzen volcano is about 99% mantle derived, while that in Aso volcano is only about 72%. The cause of this difference needs to be fully discussed. Since both volcanoes are located in the same tectonic field, the Beppu-Shimabara graben zone, the helium isotopic ratios of the source magma produced by partial melting of the upper mantle would be the same. Since there is no difference in the process of magma rising in the crust, isotopic variations would not occur here either. Thus, the variation would have occurred after the source magma entered the magma chamber just below the volcano and before the process of ejection into the central cone or flow into the surrounding subsurface hydrothermal system occurred. Even though it is not perfectly conclusive and limited because there are no volcanic gas samples taken directly from the crater, the difference in maximum values estimated by the HD model would reflect evolutions in the helium isotope ratios of the magma chamber.

U and Th concentrations in volcanic rocks ejected by recent eruptions have been reported: 2.2 ± 0.4 ppm (error of 1σ variation) and 9.1 ± 0.9 ppm (1σ) for Unzen-Fugendake²⁴ and 2.0 ± 1.0 ppm (1σ) and 7.9 ± 3.7 ppm (1σ) for Aso-Nakadake²⁵. Thus, there is not much difference in U and Th concentrations between them. Assuming that the helium-3 concentrations in

the primary magmas of both volcanoes are the same, the difference in helium isotopic ratios at the magma chamber based on the HD model is simply an age effect due to the addition of radiogenic helium in a closed system. The evolutionary curve of helium ingrowth in magmas has been calculated for systems of mid-ocean ridge basalts, and in the Holocrystalline Tholeiite, the $^3\text{He}/^4\text{He}$ ratio decreases from 8 Ra to 2 Ra in 100,000 years⁹. Assuming the reported U and Th concentrations^{24,25}, the initial ^3He concentration is 1.2×10^{-12} ccSTP/g, to be intermediate between the Holocrystalline Tholeiite and the Glassy Tholeiite, the $^3\text{He}/^4\text{He}$ ratio decreases from 8 Ra to 7.7 Ra in 10,000 years and further to 5.9 Ra in 100,000 years (Supplementary Fig. 1). If the caldera eruption of Aso volcano was about 80,000 years ago²⁶, the helium isotope ratio of the magma at that time decreases to 6.2 Ra on the evolution curve. The larger the caldera eruption, the more likely it is that magma remained in the magma reservoir even afterward²⁷. If the helium ejected from the current Nakadake cone is due to this residual magma from about 80,000 years ago, it is consistent with the estimated isotope ratio of 6.0 Ra. In other words, the assumption of no new magma supply is consistent with the results of seismic tomography described in the previous section: No mantle passage is visible below Aso volcano. On the other hand, Unzen volcano itself has an eruption history of 500,000 years²⁸, while Fugendake is younger, with a fission-track age of about 6000 years²⁹ for the lava. If the source magma was newly supplied from the mantle at this age, the decrease of helium isotope ratio is very small and can be calculated to be 7.8 Ra, slightly lower than the estimated value of 7.95 Ra. Thus, the difference in the highest helium isotope ratios ($r = 0$ at HD model) between Aso/Nakadake and Unzen/Fugendake can be explained by the age effect of magma chamber, which is consistent with the results of seismic tomography (Fig. 3).

Next, we discuss the geographic impact of volcanic activity on the surrounding geothermal water system based on helium isotope ratios. Qualitatively, this effect is seen in Fig. 2, where higher helium isotope ratios are observed at greater distances from the central cone. Quantitatively, it can be evaluated by the distance to 50% of the $r = 0$ value (the maximum $^3\text{He}/^4\text{He}$ ratio for that volcano) in the HD model curve⁹. For Aso volcano, the distance from the crater to 3.0 Ra, that is half of the maximum value of 6.0 Ra, is 21 km (hereafter we call it the half-distance). On the other hand, when the same calculation is applied to Unzen volcano, the half-distance at which the helium isotope ratio reaches 4.0 Ra is 10.2 km. Thus, the strength of mantle helium spreading from just below the volcano to the surrounding geothermal system is twice as great for Aso volcano compared to Unzen volcano. This strength is due to the higher amount of helium flux from the point source in the HD model. As mentioned above, the flux cannot be calculated for $r = 0$, so assuming $r = 1$ km, the ^3He -flux for Unzen volcano is estimated to be 1.8×10^6 atoms $\text{m}^{-2} \text{sec}^{-1}$. This value is only 31% of that of Aso volcano. Thus, Aso volcano may have more mantle helium influence on more distant hot springs than Unzen volcano.

As mentioned above, comparing Aso and Unzen volcanoes, Nakadake central cone has a lower helium isotope ratio, while the effect on the surrounding area is greater than that of Fugendake crater in terms of half-distance. Since it is impossible to make a general comparison between caldera volcanoes and stratovolcanoes from these two volcanoes only, we summarize the helium isotope ratio data published so far for the surrounding volcanoes (Table 1). Data from Aso and Unzen volcanoes in this study are compared with those from Ontake²⁰, Nevado del Ruiz³⁰, Kusatsu-Shirane³¹, and Hakone volcanoes³² (Supplementary Fig. 2). Although the data from Ontake are

Table 1 Summary of helium isotope variations around volcanoes.

| Volcano | Max. $^3\text{He}/^4\text{He}$ (Ra) | ^3He -flux ($\times 10^9 \text{ atoms m}^{-2} \text{ sec}^{-1}$) | r (1/2) (km) | Rock-type | Elevation (m) | Reference |
|------------------------|-------------------------------------|-----------------------------------------------------------------------------|--------------|-----------------|---------------|-------------------------------------------|
| <i>Strato volcano</i> | | | | | | |
| a Ontake | 7.89 | 37.3 | 15.1 | Andesite | 3063 | Takahata et al., 2003 |
| b Nevado del Ruiz | 6.73 | 9.5 | 8.3 | Andesite | 5321 | Williams et al. ³⁰ |
| c Kusatsu | 7.97 | 27.8 | 12.5 | Andesite-dacite | 2176 | Sano et al. ³¹ |
| d Unzen | 7.95 | 17.8 | 10.2 | Andesite | 1500 | This work & Morikawa et al. ¹⁴ |
| Average | 7.64 | 23.1 | 12 | | | |
| <i>Caldera volcano</i> | | | | | | |
| e Hakone | 6.99 | 53.9 | 19.1 | Bbasalt-dacite | 1438 | Sakamoto et al. ³² |
| f Aso | 6.01 | 59.3 | 22.3 | Basalt-andesite | 1592 | This work |
| Average | 6.50 | 56.6 | 20.7 | | | |

r(1/2): distance from the cone to the place where estimated Ra becomes half of the maximum.

accumulated over multiple years, we adopted the 1993 observations, which covered even the fumarolic gas in the central cone²⁰.

Hakone volcano is a caldera volcano⁵, and its explosive eruption that formed the caldera occurred 80,000–40,000 years ago³³. Since there are only two data points outside the caldera of this volcano (Supplementary Fig. 2d), the HD model fitting was tentatively made, although it is a difficult task. The estimated maximum $^3\text{He}/^4\text{He}$ ratio ($r = 0$) is 6.99 Ra, lower than that of Unzen volcano. Again, this may be attributable to helium ingrowth (Supplementary Fig. 1). Taking into account all the compiled data (Table 1), we deem that the maximum helium isotope ratios for the stratovolcanoes are higher than those for the caldera volcanoes based on the values averaged over both types. On the other hand, the helium-3 flux is larger for the caldera volcanoes, and the half-distance, which indicates the impact on the surrounding geothermal system, is also larger. In this compilation (Table 1), rock-type of stratovolcano is mainly andesite, while that of caldera volcano is basalt. This may be related to aquifer rock system and their U and Th contents, which are important parameter of HD model and $r(1/2)$. This should be discussed in future work.

A simplified schematic diagram of Unzen volcano (stratovolcanoes) and Aso volcano (caldera volcano) investigated in this study is shown in Fig. 4. In the Unzen (Fig. 4a), the magma chamber is young and supplied continuously with primordial helium from the mantle. In contrast, the horizontal and radial flux of magmatic helium is relatively small and is soon diluted by radiogenic helium in the aquifer, resulting in quick decrease in isotopic ratios. On the other hand, in Aso volcano (Fig. 4b), the magma reservoir is old and the supply from the mantle has probably ceased. Therefore, the helium isotope ratio observed at the central cone of the post-caldera volcano is lower than the upper mantle value of 8 Ra. On the other hand, the flux of magmatic helium in the radial direction is relatively large probably due to the development of the aquifer, resulting in high helium isotope ratios even in hot springs far from the central cone. It is well documented that faulted and fractured intra-caldera rocks may trap meteoric waters that are heated by subjacent magma. Because of their large size, calderas such as Yellowstone and Taupo Zone may host larger, longer lived geothermal systems than those in typical stratovolcanoes (ref. ³⁴). In conclusion, the results obtained in this study alone may not generalize differences between system of stratovolcanoes and caldera volcanoes around the world, and cannot answer the proposition that stratovolcanoes would evolve into caldera volcanoes. However, Ontake volcano, with its larger radial flux, is closer in character to a caldera volcano such as Aso than Unzen, and may be more likely to evolve to a caldera volcano after tens of thousands of years.

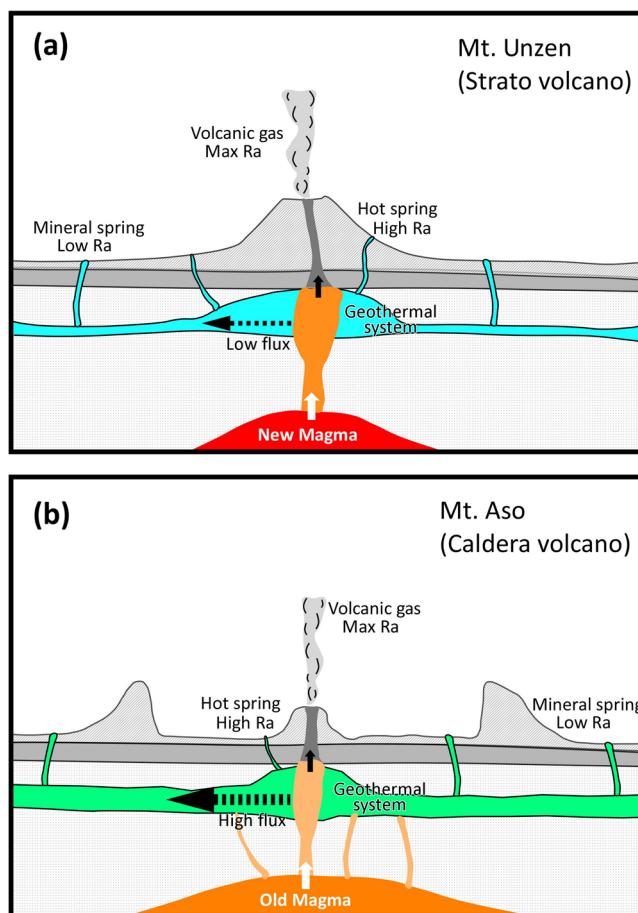


Fig. 4 Schematic diagram of the structure of a stratovolcano and a caldera volcano. Schematic diagram of the structure of **a** a stratovolcano and **b** a caldera volcano, based on the concept of helium isotope geochemistry.

Methods

Helium isotopes and gas geochemistry. Hot spring water, groundwater, and free gas were collected in Aso volcano within the caldera shown in Fig. 1, and in the Kumamoto Plain outside the caldera in addition to those in Unzen volcano. Hot spring water and groundwater were sampled using a syringe with a two-tip, and fluid samples were led into a vessel made of a special glass with low helium permeability. Hot spring free gas was collected by the water displacement method using a funnel⁹. Noble gas isotope ratios, main component chemical compositions, nitrogen and carbon isotope ratios were measured at the Atmosphere and Ocean Research Institute, University of Tokyo. Gas samples or a portion of the gas samples extracted from hot spring water were introduced into

a metallic ultra-high vacuum line, and noble gases were separated and purified with a hot titanium getter and an activated carbon trap cooled to liquid nitrogen temperature. After measuring the $^4\text{He}/^{20}\text{Ne}$ ratio using online quadrupole mass spectrometer, the helium and neon were separated by an activated carbon trap held at 40 K. The fully purified helium was introduced into a vacuum static noble gas mass spectrometer (Helix SFT, GV instrument) and the $^3\text{He}/^4\text{He}$ ratio was measured³⁵. The values obtained were normalized against laboratory standard air samples. The chemical composition of the gas samples was measured by a dynamic quadrupole mass spectrometer. The remaining gas samples were introduced into a glass high vacuum line, where nitrogen and argon were purified using a liquid nitrogen trap and hot copper oxide getter. Nitrogen was introduced into a static vacuum nitrogen isotopic mass spectrometer for measurement³⁶. Argon isotope ratios were measured by quadrupole mass spectrometer.

Seismic tomography. Three-dimensional (3-D) P- and S-wave velocity (V_p , V_s) models of the Kyushu subduction zone were determined by using 179,951 P- and 166,719 S-wave arrival times of 6002 local shallow and intermediate-depth earthquakes at depths of 0–170 km, which were recorded at 206 seismic stations in Kyushu during June 2002 to November 2017 (ref. 15). These data were selected from the JMA (Japan Meteorological Agency) unified earthquake catalogue. Theoretical travel times and ray paths are calculated using a 3-D ray tracing technique¹⁵. A 3-D grid is arranged in the crust and upper mantle under the study region with a lateral grid interval of ~20 km and vertical grid intervals of 9–20 km. V_p and V_s perturbations from a 1-D starting model at the grid nodes are taken as unknown parameters. The velocity perturbation at any point in the study volume is calculated by linearly interpolating the velocity perturbations at eight grid nodes surrounding that point. The large and sparse system of observation equations relating the arrival-time data with the unknown parameters is solved by using a least-squares method. The local earthquakes are relocated in the inversion process. As a result, 3-D V_p and V_s models of the study region are obtained, and 2-D V_p and V_s images along any profile (such as Fig. 3) can be plotted from the 3-D tomographic models.

Data availability

The data supporting the findings of this study are included in the article and Supplementary Information. The source data of Fig. 2 and Supplementary Fig. 2 are publicly available in the EarthChem data repository (<https://www.earthchem.org/>) (ref. 37).

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Author contributions

Y.S. and T.K. designed the study and analyzed the data, Y.S., T.K., M.Z., T.O., and T.S. carried out fluid sampling, N.T., A.-T.C., M.Z., and H.L. carried out isotope analysis, D.P. provided seismic tomography, Y.S., T.K., M.Z., N.T., T.O., T.S., Y.N., T.P.F., and D.P. contributed to the writing of the paper.

Competing interests

The authors declare no competing interests.

Additional information

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