Radioactive equilibria and disequilibria of U-series nuclides in the products from Izu arc volcanoes, Japan

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Summary. Activity ratios among ^{238}U – ^{230}Th – ^{226}Ra in the products from Izu arc volcanoes, Japan, were observed in order to estimate the time scale of magmatic processes and the magma generation for Izu arc volcanism. Activity ratios of $^{238}\text{U}/^{230}\text{Th}$ and $^{226}\text{Ra}/^{230}\text{Th}$ in the basaltic and andesitic products from Izu arc volcanoes were greater than unity, being enriched in ^{238}U and ^{226}Ra relative to ^{230}Th . The $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio versus $^{238}\text{U}/^{230}\text{Th}$ activity ratio diagram for these products showed positive correlation, suggesting that the ^{238}U – ^{230}Th – ^{226}Ra disequilibria occurred during the magma genesis by the additions of U- and Ra-rich fluids derived from the subducting slab by dehydration to the mantle wedge. The ^{230}Th – ^{226}Ra radioactive disequilibria observed in the basaltic and andesitic products imply a short period of time (< 8000 years) between the magma genesis and the eruption. The majority of rhyolitic products was considered to be almost in equilibrium of $^{238}\text{U} = ^{230}\text{Th} = ^{226}\text{Ra}$. The observation that ^{238}U – ^{230}Th – ^{226}Ra for the rhyolite are in radioactive equilibrium suggested that the rhyolitic magma from Izu arc was generated in the partial melting of the earth crust heated by the basaltic magma of high temperature.

1. Introduction

The decay chain of ^{238}U ($t_{1/2} = 4.5 \times 10^9$ y) contains a number of radioactive nuclides with different chemical properties and half-lives. During magmatic processes from the magma generation in the mantle to the eruption, the differences in chemical properties between parent and daughter nuclides will allow them to be fractionated, generating radioactive disequilibrium. As the time since the fractionation can be assessed using the half-lives of the daughter nuclides (e.g., $t_{1/2} = 7.5 \times 10^4$ y for ^{230}Th , and $t_{1/2} = 1.6 \times 10^3$ y for ^{226}Ra), investigations of radioactive disequilibria among U-series nuclides in volcanic rocks can potentially provide the information on the time scale of magmatic processes [1]. While the ^{238}U – ^{230}Th disequilibria can be used to estimate the time scale of an event of

the U–Th fractionation within the period of 3.5×10^5 y (i.e., 5 half-lives yield 97% of the equilibrium), the ^{230}Th – ^{226}Ra disequilibria can be used to estimate the time scale of an event of the Th–Ra fractionation within the period of 8.0×10^3 y.

The generation of arc magmas is generally inferred to be induced by the partial melting of the mantle wedge by the addition of the fluid derived from the subducting slab [2].

The majority of the arc volcanic rocks is characterized by ^{238}U -excess over ^{230}Th [3, 4]. Because U and Ra are highly mobile element in aqueous fluids relative to Th [5, 6], the excess of ^{238}U over ^{230}Th in the arc volcanic rocks has generally been ascribed to the addition of U-rich fluids derived from the subducting slab by dehydration.

The investigations of the disequilibria between ^{230}Th and ^{226}Ra in fresh volcanic rocks from many arcs have shown that the majority of arc volcanic rocks are characterized by ^{226}Ra excess over ^{230}Th [3, 4]. The ^{230}Th – ^{226}Ra disequilibria observed in the most arc volcanic rocks along with the positive correlations with $^{238}\text{U}/^{230}\text{Th}$ activity ratio and Ba/Th concentration ratio (the latter is usually regarded as a good indicator of the fluid addition) may also be attributed to the addition of U- and Ra-rich fluids derived from the subducting slab by dehydration, implying that the time scale from the magma genesis to the eruption was less than 8.0×10^3 y.

The ^{238}U – ^{230}Th disequilibria in the products from Izu arc volcanoes have previously been observed [7–11]. The observations showed that while the $^{230}\text{Th}/^{238}\text{U}$ activity ratios of basaltic and andesitic products were smaller than unity, being enriched in ^{238}U relative to ^{230}Th [7–10], the $^{230}\text{Th}/^{238}\text{U}$ activity ratios of rhyolitic products were considered to be almost unity [11].

As the prior studies of radioactive disequilibrium between ^{230}Th and ^{226}Ra in the basaltic products from Izu arc volcanoes were limited [12, 13], although Miyake-jima volcano has been intensively investigated [7, 8], ^{226}Ra in the basaltic products from Fuji and Izu-Oshima volcanoes located along Izu arc are presently measured.

This report deals with the discussion including the latest observations of the $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio in the basaltic products from Fuji and Izu-Oshima volcanoes, which belong to Izu arc in order to estimate the time scale of magmatic

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Fig. 1. Locality map of Izu arc volcanoes, Japan.

processes. Discussion will extend to the comparison with the activity ratio of the rhyolitic products [14] from Izu arc volcanoes.

2. Samples and analytical methods

Samples

Observations were carried out with 16 volcanic rock samples from Fuji (800–802 AD, 864–866 AD, 937 AD and 1033 AD) and Izu-Oshima (9th C, 1338 AD, 1421 AD, 1552 AD, 1684 AD, 1777–1778 AD and 1950–1951 AD) volcanoes located along Izu arc, Japan. For 6 volcanic rock

samples of these samples, U and Th isotopes were also measured (Table 1). Locality map is shown in Fig. 1.

Analytical methods

SiO₂ concentration in the volcanic rock samples was determined by X-ray fluorescence spectrometry (XRF) using fused glass beads made by mixing sample with anhydrous lithium tetraborate as a flux. The experimental procedures have been described in detail by Nakayama and Nakamura [15].

Determination of U and Th isotopes in the volcanic rock sample was made by an isotope dilution analysis coupled with α -ray spectrometry. Uranium-232 was spiked for determination of U isotopes and commercially available Th reagent having different ²³⁰Th/²³²Th activity ratio from those in the volcanic rock samples was spiked for determination of Th isotopes. Detailed chemical separation and purification procedures of U and Th isotopes from the lava samples have previously been described in Takahashi *et al.* [16]. The purified U and Th fractions were electrodeposited onto stainless steel planchets of 1 inch $\varnothing$ by Lee's method [17] for α -ray counting, respectively.

Determination of ²²⁶Ra in the volcanic rock samples was made by non-destructive γ -ray spectrometry. About 100 g of volcanic rock sample was packed into a canister of tin-can (76 mm $\varnothing$, 24 mm high). The canister was sealed with epoxy resin (Araldite®) completely. The volcanic rock sample for the γ -ray measurement thus prepared was stored for 1 month in order that ²¹⁴Pb reached the equilibrium with ²²⁶Ra within the canister.

Table 1. SiO₂ contents, U–Th–Ra data for the historical eruption products from Fuji and Izu-Oshima volcanoes.

Volcano (Eruption year)	Sample identification	SiO ₂ [%]	Content		²²⁶ Ra [fg · g ⁻¹]	²³⁴ U/ ²³⁸ U	Activity ratio		
			U [μg · g ⁻¹]	Th			²³⁸ U/ ²³² Th	²³⁰ Th/ ²³² Th	²²⁶ Ra/ ²³⁰ Th
Fuji									
(800–802 AD)	Tak-1 96080901 ^a	49.3 (0.1) ^b	0.49 ± 0.02	1.17 ± 0.06	220 ± 6	1.01 ± 0.02	1.30 ± 0.08	1.09 ± 0.02	1.56 ± 0.09
(864–866 AD)	Ao-95122802 ^a	49.7 (0.1) ^b	0.55 ± 0.02	1.33 ± 0.09	212 ± 6	0.98 ± 0.02	1.24 ± 0.09	1.06 ± 0.03	1.36 ± 0.11
	Ao-95120814 ^c	–	0.58 ± 0.02	1.34 ± 0.04	229 ± 6	0.98 ± 0.02	1.31 ± 0.06	1.06 ± 0.01	1.46 ± 0.06
	Ao-95101905 ^c	–	0.56 ± 0.02	1.48 ± 0.05	224 ± 7	1.02 ± 0.02	1.18 ± 0.06	1.04 ± 0.01	1.32 ± 0.06
	Ao-95120811 ^c	–	0.58 ± 0.03	1.36 ± 0.08	221 ± 9	0.99 ± 0.02	1.29 ± 0.09	1.06 ± 0.02	1.38 ± 0.10
	Ao-95060308 ^c	–	0.49 ± 0.02	1.30 ± 0.05	206 ± 5	1.01 ± 0.02	1.18 ± 0.06	1.04 ± 0.01	1.38 ± 0.06
(937 AD)	Ken-6 96080908 ^a	49.8 (0.1) ^b	0.43 ± 0.02	1.14 ± 0.05	176 ± 4	1.00 ± 0.02	1.16 ± 0.07	1.08 ± 0.02	1.29 ± 0.07
(1033 AD)	Ken-5 96080907 ^c	50.3 (0.1) ^b	0.44 ± 0.01	1.08 ± 0.06	199 ± 6	0.98 ± 0.02	1.15 ± 0.07	1.06 ± 0.02	1.60 ± 0.10
Izu-Oshima									
(9th C)	OSM06060401 ^a	50.8 (0.1) ^b	0.18 ± 0.00	0.28 ± 0.01	64 ± 3	1.01 ± 0.01	1.99 ± 0.10	1.53 ± 0.03	1.36 ± 0.10
(1338 AD)	JS820626 ^a	52.1 (0.1) ^b	0.20 ± 0.01	0.40 ± 0.02	73 ± 3	1.00 ± 0.02	1.52 ± 0.08	1.24 ± 0.02	1.31 ± 0.09
(1421 AD)	OSM06060301 ^a	52.1 (0.5) ^b	0.19 ± 0.01	0.34 ± 0.01	86 ± 6	1.01 ± 0.02	1.79 ± 0.08	1.36 ± 0.02	1.68 ± 0.13
(1552 AD)	OSM06060302 ^a	51.9 (0.5) ^b	0.20 ± 0.01	0.31 ± 0.01	77 ± 3	1.00 ± 0.02	1.99 ± 0.09	1.41 ± 0.02	1.63 ± 0.10
(1684 AD)	OSM06060303 ^a	51.5 (0.1) ^b	0.21 ± 0.01	0.32 ± 0.01	60 ± 3	1.01 ± 0.02	2.00 ± 0.10	1.35 ± 0.02	1.25 ± 0.08
(1777–1778 AD)	OSM05051701 ^a	52.5 (0.1) ^b	0.18 ± 0.01	0.33 ± 0.02	85 ± 3	0.99 ± 0.02	1.61 ± 0.11	1.38 ± 0.05	1.61 ± 0.11
	JS820627-02 ^c	50.2 (0.3) ^b	0.16 ± 0.01	0.32 ± 0.02	78 ± 5	1.00 ± 0.03	1.55 ± 0.10	1.38 ± 0.03	1.58 ± 0.14
(1950–1951 AD)	OSM05051601 ^a	51.7 (0.4) ^b	0.18 ± 0.01	0.26 ± 0.02	86 ± 5	1.01 ± 0.03	2.21 ± 0.16	1.41 ± 0.02	2.14 ± 0.19

a: Uranium and Th data are taken from Table 2 of [9];

b: Relative standard deviation / %, $n = 5$;

c: Uranium and Th data are obtained in this work.

Radioactivity measurement

Activity ratios of U and Th isotopes were obtained by α -ray spectrometry using a 900 mm² PIPS detector (CANBERRA, A 900-25 AB) coupled with an MCA system (Laboratory Equipment; ADC 2201A). Radioactivities of U and Th isotopes were determined by the following α -rays: 4.151 MeV and 4.198 MeV for ²³⁸U, 4.722 MeV and 4.775 MeV for ²³⁴U, 5.263 MeV and 5.320 MeV for ²³²U, 3.954 MeV and 4.013 MeV for ²³²Th, and 4.621 MeV and 4.688 MeV for ²³⁰Th, respectively.

Gamma-ray intensity was measured with an HPGe semiconductor detector (Princeton Gamma-Tec, IGC-10200 NPR) coupled with an MCA system (Aptec Engineering, 5008). Radioactivity of ²²⁶Ra was determined by the 351.9 keV γ -ray from ²¹⁴Pb in equilibrium with ²²⁶Ra.

3. Results and discussion

The present determination procedures were applied to JB-1, the geochemical reference material of basalt, issued by Geological Survey of Japan (The National Institute of Advanced Industrial Science and Technology). The reference material should be in radioactive equilibrium among U-series nuclides, because the eruption age of JB-1 is 7.59 Ma [18]. The activity ratios among ²³⁸U–²³⁴U–²³⁰Th–²²⁶Ra in JB-1 were observed to be unity.

Table 1 lists the contents of U, Th (in $\mu\text{g g}^{-1}$) and ²²⁶Ra (in fg g^{-1}) along with SiO₂ content (in %), and the activity ratios of ²³⁴U/²³⁸U, ²³⁸U/²³²Th, ²³⁰Th/²³²Th and ²²⁶Ra/²³⁰Th in the historical eruption products from Fuji and Izu-Oshima volcanoes.

As ²³⁴U in the products has been reported to be in equilibrium with ²³⁸U [9], ²³⁰Th in the products can be regarded as the direct decay product of ²³⁸U.

Radioactive equilibria and disequilibria among ²³⁸U–²³⁰Th–²²⁶Ra in the volcanic rock samples

Fig. 2 is the (²³⁰Th/²³²Th)–(²³⁸U/²³²Th) activity ratio diagram for the products from Izu arc volcanoes [9, 11].

While the majority of rhyolitic products was almost in radioactive equilibrium [11], the activity ratios of basaltic and andesitic products from Izu arc showed ²³⁰Th/²³⁸U < 1 [9]. The suite of (²³⁰Th/²³²Th)–(²³⁸U/²³²Th) data for the basaltic and andesitic products [9] forms a broad inclined array which is similar to the data sets for Tonga-Kermadec [19] and Mariana [20] island arcs.

Fig. 3 is the (²²⁶Ra/²³⁰Th)₀ activity ratio versus ²³⁸U/²³⁰Th activity ratio diagram for the products [12–14]. (²²⁶Ra/²³⁰Th)₀ represents the decay-corrected ²²⁶Ra/²³⁰Th value for ²²⁶Ra to the eruption year.

The ²³⁸U/²³⁰Th and (²²⁶Ra/²³⁰Th)₀ activity ratios of the products show a bimodal distribution: one is the basalt and andesite group and the other is the rhyolite group [14]. The majority of rhyolitic products converges on the radioactive equilibrium point of ²³⁰Th = ²²⁶Ra and ²³⁸U = ²³⁰Th. The observation that ²³⁸U–²³⁰Th–²²⁶Ra for the rhyolite are in radioactive equilibrium suggested that the rhyolitic magma from Izu arc was generated in the partial melting of the

Fig. 2. ²³⁰Th/²³²Th activity ratio versus ²³⁸U/²³²Th activity ratio diagram for the products from Izu arc volcano. Data for basaltic and andesitic products (from Fuji, Izu-Oshima, Miyake-jima and Hachijo-jima volcanoes) are taken from [9], and those for rhyolitic products (from Niiijima, Kozu-shima and Higashi Izu volcanoes) are taken from [11].

Fig. 3. (²²⁶Ra/²³⁰Th)₀–(²³⁸U/²³⁰Th) activity ratio diagram for the products from Izu arc volcanoes. Data for rhyolitic products (from Niiijima, Kozu-shima and Higashi Izu volcanoes) are taken from [14]. Data for basaltic and andesitic products (from the 1707 eruption of Fuji volcano, and from the 1986 eruption of Izu-Oshima volcano) from [12] and [13] are included.

earth crust, where ²³⁸U–²³⁰Th and ²³⁰Th–²²⁶Ra are in equilibrium, heated by the basaltic magma of high temperature [21].

Fig. 3 shows positive correlations between ²³⁸U/²³⁰Th activity ratio and (²²⁶Ra/²³⁰Th)₀ activity ratio in the basaltic and andesitic products. The correlation suggests that the ²²⁶Ra excess over ²³⁰Th in the products may also be resulted by the addition of U- and Ra-rich fluids derived from the subducting slab by dehydration. Assuming that ²²⁶Ra excess over ²³⁰Th was generated at the magma genesis, the time scale from the magma genesis to the eruption, including the melting of the mantle wedge, magma storage and magmatic differentiation from basalt to andesite is less than 8.0×10^3 y.

4. Conclusion

Basaltic magmas from historical eruptions of Fuji and Izu-Oshima volcanoes showed $^{230}\text{Th} < ^{238}\text{U}$ and $^{226}\text{Ra} > ^{230}\text{Th}$ disequilibria which is common characteristic to the basaltic and andesitic magmas from Izu arc volcanoes. The $^{226}\text{Ra} > ^{230}\text{Th}$ disequilibrium observed in the magma implies that the time scale from the magma genesis to the eruption is less than 8000 y.

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