

Unzen Volcano : the 1900-1992 eruption

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12. Geochemical Study of Unzen Volcano by Noble Gas Measurement

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Introduction

Recent results of He isotope analyses on various terrestrial samples such as fumarolic gases (e.g., Nagao et al., 1981), Mid Oceanic Ridge basalts (e.g., Lupton et al., 1975) and mantle derived minerals (e.g., Kaneoka and Takaoka, 1980) indicate that the $^3\text{He}/^4\text{He}$ ratio in samples derived from deep interiors of the earth is an order of magnitude higher than the atmospheric ratio. The high $^3\text{He}/^4\text{He}$ ratio has been attributed to emission of primordial He which had been trapped in the earth's interior with a high $^3\text{He}/^4\text{He}$ ratio (e.g., 1.4×10^{-4}) at the early stage of the earth formation (Clarke et al., 1969; Mamyrin et al., 1969).

Noble gases are chemically inert, and the elemental and isotopic compositions are not affected by chemical reactions between volcanic gases and surrounding rocks of volcanic conduits. The scarcity in the atmosphere and large differences in the isotopic and elemental ratios between the magmatic and the atmospheric components make it easy to detect a small change in the noble gas composition of volcanic emanations resulting from the change in magmatic activity.

Figure 12-1 compares the $^3\text{He}/^4\text{He}$, $^{40}\text{Ar}/^{36}\text{Ar}$ and $^4\text{He}/^{20}\text{Ne}$ ratios for Japanese fumarolic gases with the atmospheric ratios. The $^3\text{He}/^4\text{He}$ ratio of fumarolic He is about 7 times higher than the atmospheric ratio. The fumarolic $^{40}\text{Ar}/^{36}\text{Ar}$ ratio is variable from 310 to 315 (Nagao et al., 1979, 1981). These low ratios for fumarolic Ar are assigned to contamination by atmospheric Ar which dissolved in fumarolic gases probably through groundwater. The $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 330-356 has been reported for trapped Ar in large phenocrysts separated from modern lavas (Marty

et al., 1989; Takaoka, 1989). If large phenocrysts solidified in magma chambers and trapped ambient Ar, these values represent the magmatic $^{40}\text{Ar}/^{36}\text{Ar}$ ratio, which is believed to be considerably higher than the ratio for fumarolic Ar. The isotopic ratios of Ne, Kr and Xe, and $^{38}\text{Ar}/^{36}\text{Ar}$ for fumarolic gases are identical with the atmospheric ones. The $^4\text{He}/^{20}\text{Ne}$ ratio for the fumarolic gas is more than three orders of magnitude higher than that for air and air-saturated water (Nagao et al., 1979, 1980, 1981; Marty et al., 1989).

These results suggest that by measuring the noble gas composition in fumarolic and hot-spring gases, we can get useful information on the magmatic activity of a volcano and it may be helpful for eruption prediction: With the elevated activity, the noble gas composition would change toward the magmatic one and as the activity reduces, the composition would resemble the atmospheric value found for air and air-saturated water because admixture of groundwater into fumarolic gases increases with the reduction of

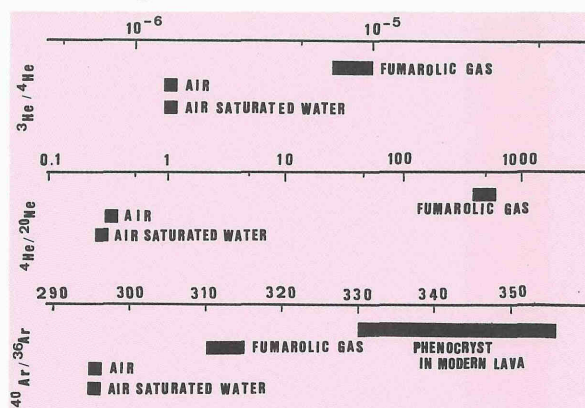


Fig. 12-1. $^3\text{He}/^4\text{He}$, $^4\text{He}/^{20}\text{Ne}$, $^{40}\text{Ar}/^{36}\text{Ar}$ ratios for fumarolic gases, large phenocrysts in modern lavas, and atmospheric component (air and air-saturated water).

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magmatic gas pressure. Nagao et al. (1980) have reported that noble gas compositions in fumarolic gases at Showa-shinzan Volcano changed toward the atmospheric ones as the magmatic activity decreased from 1958 to 1977. Sano et al. (1991) have found an anomalous increase in the $^3\text{He}/^4\text{He}$ ratio prior to the explosive eruption of Mt. Mihara, at a steam well located 3 km from the volcano. In this study, we have analyzed gas samples collected at the summit fumarole (Kujukushima) of Unzen Volcano, Shimabara Spa, Unzen Spa, and Karimizu Spa. Analyses were carried out for the elemental abundances of five stable noble gases and the isotopic ratios of He, Ne and Ar, in order to know whether the eruption of Unzen Volcano caused any changes in the noble gas composition for emanations at the hot springs or not.

Experimental

Shimabara Spa is located 7 km east from Unzen Volcano. Free gas bubbling at an underground pool (at 26°C) of the Shimabara Kanko Hotel was collected. The water and gas are flowing from an unused bore (300 m depth) of the hotel. The gas is CO_2 and the water is saturated with it (Ohta, 1973). Unzen Spa is located 4 km southwest from the volcano. There are many fumarolic vents of various geothermal activities in this area. Free gas gushing in a small pond (at 50 to 80°C, depending on atmospheric temperature) at Sheishichi Jigoku was collected. Karimizu Spa is a small pond, located at 9 km far from the Volcano. Large amounts of CO_2 gas are gushing in warm water (at 26°C). The gas samples were collected approximately once a month in glass containers with stop cocks at both ends by means of displacing water by sample gases (Nagao et al., 1981). The sample gases were analyzed within six months after collection. Diffusive loss of helium through glass wall is less than 3% during storage, and therefore, the change of isotopic ratio is negligible.

The samples were analyzed with a VG-5400 mass spectrometer at the Institute for Study of the Earth's Interior, Okayama University. Analytical procedures used in this work are similar to those used by Nagao et al. (1981). The

mass spectrometer was calibrated for noble gas sensitivities and correction factors for mass discrimination by measuring the standard gases prepared from known amounts of air. Corrections for blank and doubly-charged ^{40}Ar and CO_2 were negligible.

Results

The isotopic ratios of He, Ne and Ar, the abundance of He and the relative abundances of five stable noble gases are summarized in Table 12-1, together with sampling dates. The atmospheric data are also given for comparison. Errors attached to isotopic ratios are statistical ones and correspond to 68 % confidence, and errors for the elemental abundances, are about 10 %.

At Shimabara Spa, the $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios decreased after the first lava dome erupted on 20 May 1991, as found in Fig. 12-2, while the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio was kept constant. The $^4\text{He}/^{20}\text{Ne}$ ratio increased anomalously before the lava eruption and then decreased to the earlier value after that. The elemental abundances relative to ^{36}Ar also decreased after the lava eruption. These results of the temporal variations of noble gas compositions indicate that after the lava eruption, the proportion of the atmospheric component increased, and suggest a possible effect of the volcanic activity at Unzen Volcano on the noble gas composition in the bubbling gas at Shimabara Spa.

At Unzen Spa, the variation of isotopic ratios is not simple. The noble gas composition indicates that samples U-8, U-9 and U-11 were contaminated by the atmospheric gases and in addition, samples U-9 and U-11 were affected by isotope fractionation because the $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{38}\text{Ar}/^{36}\text{Ar}$ ratios are lower than the atmospheric ratios, as found in Table 12-1. The isotope fractionation occurred underground probably in migration of fumarolic gases. The isotopic trend found for isotope-fractionated Ne and Ar is also complicated because light isotopes are enhanced for $^{38}\text{Ar}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios, whereas heavy isotopes are enhanced for $^{20}\text{Ne}/^{22}\text{Ne}$ and $^4\text{He}/^{20}\text{Ne}$ in samples U-9 and U-11. Because of the large isotope fractionation effect, it is difficult to find the magmatic effect from Unzen Volcano

on the noble gas composition in the samples collected at the present site of Unzen Spa. However, they can give useful data on the behavior of isotopic signatures of noble gases which migrate underground to the surface. This interesting issue will be reported elsewhere.

At Karimizu Spa, both elemental and isotopic ratios of He, Ne and Ar are practically constant for all samples with a few exceptions. We can use the samples from this site for reference samples to know analytical precisions.

Discussion

Since the magmatic $^3\text{He}/^4\text{He}$, $^{40}\text{Ar}/^{36}\text{Ar}$ and $^4\text{He}/^{20}\text{Ne}$ ratios are considerably higher than the atmospheric ones, as noted earlier, the elevation of these ratios indicates an increase of the magmatic component, while the reduction does an increase of the atmospheric component in fumarolic and hot-spring gases. From this point of view, the decrease in the He and Ar isotopic ratios for the samples collected after the lava eruption can be explained by a decrease in the

proportion of the magmatic component to the atmospheric one at Shimabara Spa.

It is supposed that pore pressure was highly elevated before the lava eruption because magma pushed aside basement rocks and volatiles which were produced from the magma and by magmatic heating of groundwater were also accumulated to increase the pressure without open conduits. Considerable amounts of bulge of the mountain body which have been detected prior to the lava eruption indicate the huge elevation of pressure. This should enhance the admixture of magmatic component into aquifers through fissures. After the eruption, the pressure was relaxed slowly and the admixture of magmatic component should be reduced, and consequently the proportion of the atmospheric component should increase. The decrease in the $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios for sample S-5, which was collected 13 days after the appearance of the first lava dome, can be explained by this hypothesis. The variation of the elemental ratio such as $^3\text{He}/^{36}\text{Ar}$ also support it.

Another, possible explanation is that the isotopic variation resulted from an increase in the proportion of the atmospheric component

Table 1 Isotopic and elemental compositions of noble gases.

Sample No.	Sampling date	$^3\text{He}/^4\text{He}$ ($\times 10^{-6}$)	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{38}\text{Ar}/^{36}\text{Ar}$	$^{40}\text{Ar}/^{36}\text{Ar}$	$^4\text{He}/^{20}\text{Ne}$	$^4\text{He}/^{36}\text{Ar}$	$^{20}\text{Ne}/^{36}\text{Ar}$	$^{84}\text{Kr}/^{36}\text{Ar}$	$^{132}\text{Xe}/^{36}\text{Ar}$	C (^4He) (ppm)
Sampling site: Kujukushima Fumarole												
T-1	03/12/1990	10.24 ± 0.12	—	—	0.187 ± 0.001	309.0 ± 0.4	395	74.2	0.202	0.0338	0.00216	21
Sampling site: Shimabara Spa												
S-1	20/03/1991	9.65 ± 0.10	9.81 ± 0.01	0.0298 ± 0.0015	0.189 ± 0.003	310.2 ± 1.1	167	43.6	0.259	0.0368	0.00217	76.0
S-2	15/04/1991	9.72 ± 0.10	9.83 ± 0.01	0.0286 ± 0.0020	0.189 ± 0.003	310.2 ± 1.2	163	42.0	0.257	0.0371	0.00223	64.7
S-3	21/04/1991	9.70 ± 0.10	9.80 ± 0.02	0.0289 ± 0.0012	0.187 ± 0.003	310.9 ± 0.9	170	44.3	0.261	0.0366	0.00217	70.5
S-4	12/05/1991	9.71 ± 0.10	9.80 ± 0.05	0.0290 ± 0.0049	0.189 ± 0.003	311.7 ± 0.9	220	60.8	0.277	0.0374	0.00223	88.7
S-5	02/06/1991	9.50 ± 0.24	9.79 ± 0.04	0.0290 ± 0.0001	0.186 ± 0.002	305.7 ± 1.7	174	33.3	0.191	0.0340	0.00189	75.0
S-6	24/07/1991	9.29 ± 0.23	9.79 ± 0.04	0.0289 ± 0.0001	0.186 ± 0.003	305.0 ± 2.0	170	28.3	0.167	0.0328	0.00180	60.7
S-7	07/08/1991	9.38 ± 0.24	9.81 ± 0.04	0.0290 ± 0.0001	0.186 ± 0.002	305.9 ± 1.7	171	29.7	0.174	0.0337	0.00188	60.0
S-8	24/08/1991	9.34 ± 0.23	9.79 ± 0.04	0.0289 ± 0.0001	0.187 ± 0.004	306.2 ± 2.0	176	36.6	0.174	0.0338	0.00186	52.5
S-9	22/09/1991	9.32 ± 0.24	9.78 ± 0.04	0.0290 ± 0.0001	0.187 ± 0.003	304.8 ± 1.9	148	26.5	0.179	0.0339	0.00189	68.3
S-10	10/10/1991	9.24 ± 0.24	9.78 ± 0.04	0.0290 ± 0.0001	0.187 ± 0.002	305.9 ± 1.6	153	31.8	0.207	0.0354	0.00206	75.0
S-11	27/10/1991	9.27 ± 0.23	9.78 ± 0.04	0.0290 ± 0.0001	0.186 ± 0.002	305.6 ± 1.7	142	24.9	0.176	0.0328	0.00149	74.0
S-12	10/11/1991	9.39 ± 0.22	9.80 ± 0.04	0.0290 ± 0.0001	0.185 ± 0.002	304.1 ± 1.9	138	29.4	0.212	0.0324	0.00177	71.4

(continued)

Table 1 (continued)

Sample No.	Sampling date	$^3\text{He}/^4\text{He}$ ($\times 10^{-6}$)	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{38}\text{Ar}/^{36}\text{Ar}$	$^{40}\text{Ar}/^{36}\text{Ar}$	$^4\text{He}/^{20}\text{Ne}$	$^4\text{He}/^{36}\text{Ar}$	$^{20}\text{Ne}/^{36}\text{Ar}$	$^{84}\text{Kr}/^{36}\text{Ar}$	$^{132}\text{Xe}/^{36}\text{Ar}$	C (^4He) (ppm)
Sampling site: Unzen Spa												
UZ	03/12/1990	7.12 ± 0.12	—	—	0.188 ± 0.001	308.5 ± 0.5	170	41.7	0.234	0.0298	0.0148	16
U-1	20/03/1991	7.10 ± 0.07	9.85 ± 0.02	0.0284 ± 0.0025	0.189 ± 0.003	309.8 ± 1.1	136	36.4	0.267	0.0330	0.00261	16.5
U-2	15/04/1991	7.13 ± 0.08	9.84 ± 0.01	0.0287 ± 0.0021	0.188 ± 0.003	307.8 ± 0.8	129	37.1	0.288	0.0339	0.00184	17.6
U-3	21/04/1991	7.06 ± 0.08	9.83 ± 0.01	0.0298 ± 0.0012	0.188 ± 0.003	304.1 ± 1.0	93.1	25.7	0.276	0.0336	0.00180	20.5
U-4	12/05/1991	7.10 ± 0.07	9.82 ± 0.02	0.0294 ± 0.0018	0.190 ± 0.004	307.5 ± 0.8	123	32.7	0.249	0.0351	0.00190	19.1
U-5	02/06/1991	7.13 ± 0.17	9.80 ± 0.04	0.0290 ± 0.0002	0.187 ± 0.002	304.7 ± 1.7	98.4	21.6	0.219	0.0354	0.00204	19.2
U-6	24/07/1991	7.30 ± 0.20	9.96 ± 0.04	0.0295 ± 0.0003	0.187 ± 0.002	307.6 ± 1.7	141	12.1	0.086	0.0341	0.00186	16.2
U-7	07/08/1991	6.91 ± 0.17	9.90 ± 0.04	0.0292 ± 0.0002	0.189 ± 0.005	308.1 ± 1.9	154	38.8	0.253	0.0321	0.00181	16.0
U-8	24/08/1991	6.52 ± 0.19	9.72 ± 0.04	0.0294 ± 0.0001	0.188 ± 0.003	296.3 ± 1.0	3.14	1.74	0.554	0.0208	0.00078	13.7
U-9	22/09/1991	6.86 ± 0.21	9.68 ± 0.04	0.0293 ± 0.0001	0.185 ± 0.002	290.9 ± 1.6	4.46	1.67	0.373	0.0193	0.00065	14.6
U-10	10/10/1991	7.36 ± 0.25	9.74 ± 0.04	0.0288 ± 0.0002	0.187 ± 0.003	304.3 ± 2.0	81.9	19.6	0.239	0.0353	0.00214	22.8
U-11	27/10/1991	6.84 ± 0.18	9.68 ± 0.04	0.0290 ± 0.0001	0.185 ± 0.002	291.6 ± 1.7	7.15	2.58	0.361	0.0201	0.00071	15.1
U-12	10/11/1991	7.11 ± 0.19	9.84 ± 0.05	0.0290 ± 0.0002	0.186 ± 0.002	305.5 ± 1.6	125	27.4	0.220	0.0347	0.00189	18.0

(continued)

Table 1 (continued)

Sample No.	Sampling date	$^3\text{He}/^4\text{He}$ ($\times 10^{-6}$)	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{38}\text{Ar}/^{36}\text{Ar}$	$^{40}\text{Ar}/^{36}\text{Ar}$	$^4\text{He}/^{20}\text{Ne}$	$^4\text{He}/^{36}\text{Ar}$	$^{20}\text{Ne}/^{36}\text{Ar}$	$^{84}\text{Kr}/^{36}\text{Ar}$	$^{132}\text{Xe}/^{36}\text{Ar}$	C (^4He) (ppm)
Sampling site: Karimizu Spa												
K-1	15/04/1991	5.88 ± 0.06	9.79 ± 0.01	0.0287 ± 0.0015	0.189 ± 0.003	303.6 ± 0.9	103	22.6	0.220	0.0290	0.00213	20.5
K-2	21/04/1991	5.85 ± 0.06	9.80 ± 0.01	0.0294 ± 0.0014	0.189 ± 0.003	303.7 ± 0.8	99.6	21.5	0.215	0.0366	0.00216	20.3
K-3	12/05/1991	5.90 ± 0.07	9.81 ± 0.01	0.0288 ± 0.0008	0.188 ± 0.004	303.4 ± 1.5	101	19.1	0.190	0.0367	0.00210	19.8
K-4	02/06/1991	5.82 ± 0.15	9.80 ± 0.04	0.0289 ± 0.0001	0.188 ± 0.004	303.0 ± 1.2	99.8	17.5	0.175	0.0365	0.00212	18.5
K-5	24/07/1991	5.78 ± 0.16	9.77 ± 0.04	0.0290 ± 0.0002	0.187 ± 0.003	303.2 ± 1.4	62.6	15.5	0.249	0.0342	0.00210	17.9
K-6	07/08/1991	5.69 ± 0.14	9.80 ± 0.04	0.0290 ± 0.0001	0.187 ± 0.003	302.4 ± 1.4	100	18.0	0.179	0.0367	0.00221	20.7
K-7	24/08/1991	5.79 ± 0.16	9.85 ± 0.04	0.0289 ± 0.0002	0.189 ± 0.006	301.9 ± 3.5	109	19.1	0.176	0.0366	0.00225	20.5
K-8	22/09/1991	5.82 ± 0.16	9.83 ± 0.04	0.0291 ± 0.0001	0.187 ± 0.003	303.3 ± 1.2	86.7	16.5	0.190	0.0362	0.00215	21.6
K-9	10/10/1991	5.73 ± 0.16	9.81 ± 0.04	0.0290 ± 0.0001	0.188 ± 0.003	302.8 ± 1.5	84.0	15.1	0.180	0.0361	0.00212	22.5
K-10	27/10/1991	5.79 ± 0.15	9.78 ± 0.04	0.0290 ± 0.0001	0.187 ± 0.004	302.9 ± 1.3	80.5	16.7	0.207	0.0355	0.00204	21.6
K-11	10/11/1991	5.90 ± 0.17	9.85 ± 0.04	0.0290 ± 0.0002	0.188 ± 0.003	303.4 ± 1.2	99.5	17.1	0.172	0.0377	0.00232	27.0
Atmosphere		1.4 ¹⁾	9.80 ²⁾	0.0290 ²⁾	0.188 ³⁾	295.5 ³⁾	0.32 ⁴⁾	0.166 ⁴⁾	0.524 ⁴⁾	0.0206 ⁴⁾	0.00074 ⁴⁾	5.24 ⁴⁾

1) Mamyrin et al. (1970)

2) Eberherdt et al. (1965)

3) Nier (1950)

4) Ozima and podosek (1983)

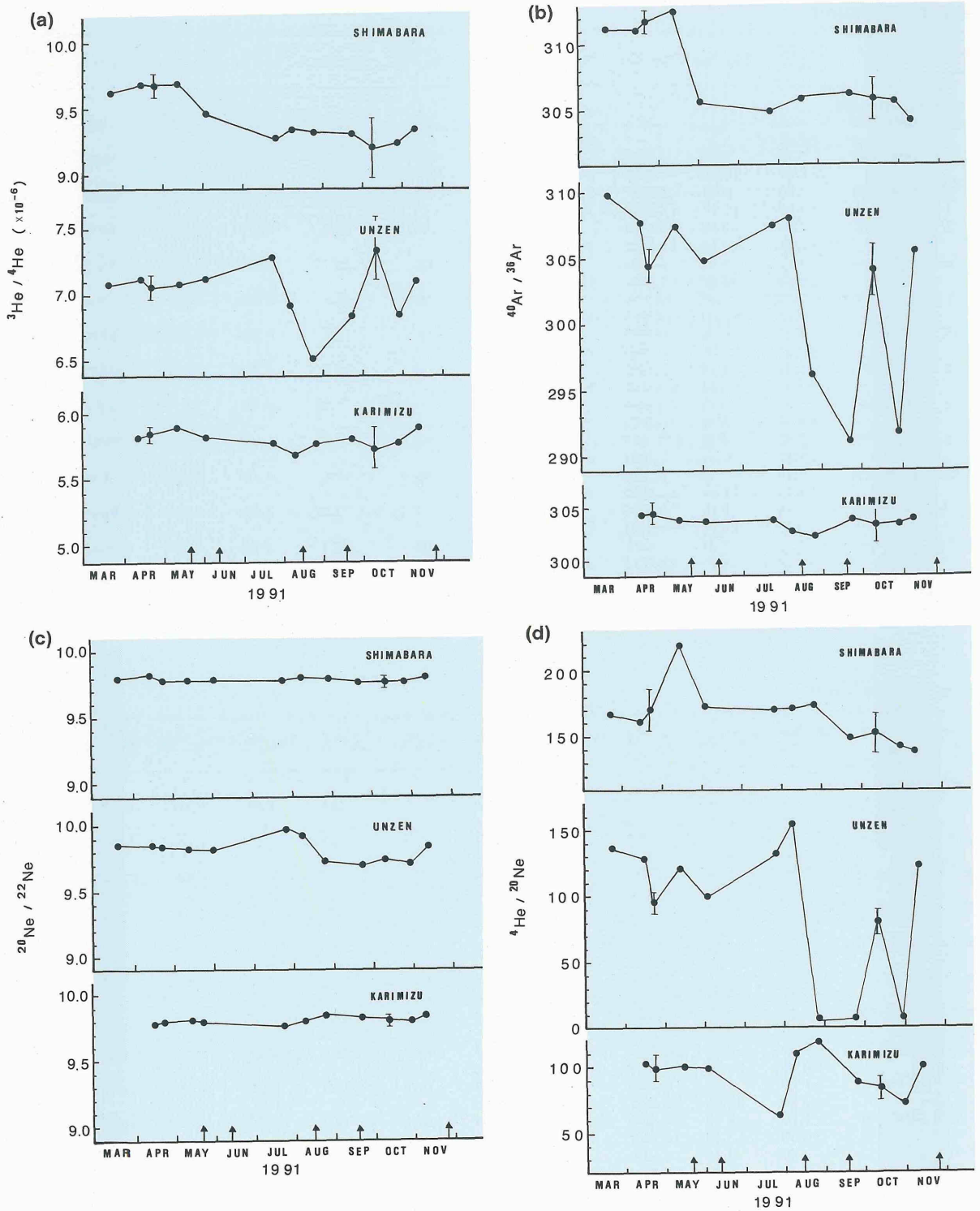


Fig. 12-2. Temporal variations of $^3\text{He}/^4\text{He}$, $^{20}\text{Ne}/^{22}\text{Ne}$, $^{40}\text{Ar}/^{36}\text{Ar}$ and $^4\text{He}/^{20}\text{Ne}$ ratios determined for gas samples collected at Shimabara Spa, Unzen Spa and Karimizu Spa. Arrows indicate eruptions of lava domes.

through groundwater caused by a seasonal change of precipitation. In this case, the observed variation is to be an accidental coincidence with the eruption. But this is not favorable because such variation has not been observed at Karimizu Spa. Considering a causality with the eruption, the observed change is a rapid response which requires a fast propagation of more than 540 m/day and thus a large driving force. The magma activity accompanied by the bulge of the volcano could give such force that drives the magmatic gases to the aquifers, and the relaxation of pressure by the lava eruption should diminish the admixture of the magmatic component rapidly.

Another question is whether the magmatic component found at Shimabara Spa originated from the same source as that for Unzen Volcano. In Fig. 12-3, the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio is plotted against the $^4\text{He}/^{36}\text{Ar}$ ratio. The bubble gases at Shimabara give a correlation line which can be regarded as a mixing line between the atmospheric component and the magmatic one. The magmatic component should be plotted on an extension of the correlation line. The gas samples from Unzen

and Karimizu are also plotted along this line except a few samples affected by the isotope fractionation. However, the sample collected at the summit vent of the volcano is plotted far from the line, as found in Fig. 12-3. The large shift of the summit sample may be a result from an elemental fractionation in which the point shifts to the right, because ^4He is enriched relative to ^{36}Ar . In this case, the magmatic component for both summit and hot-spring gases may originate from the same source. However, another view that the summit gas was not affected by the elemental fractionation is also possible. If this is the case, the magmatic component for the summit gas is different in the noble gas composition and therefore in the source from that for Shimabara Spa, because the magmatic component for the summit gas should be plotted on a mixing line passing the atmospheric point (A) and the summit gas (K) in Fig. 12-3. More complicated scenarios are also possible. Anyhow, data are not enough to specify the source of the magmatic gas found at Shimabara Spa, since we have data for only one summit sample which was collected at the early stage of volcanic activity before the lava eruption. Useful information on the relevant question will be given by analyzing fumarolic gases after the lava eruption in addition to bubble gases collected after November 1991. Measurements of Ar isotopic ratios in gaseous inclusions of the lavas will give an important constraint on the magmatic gas for this active volcano.

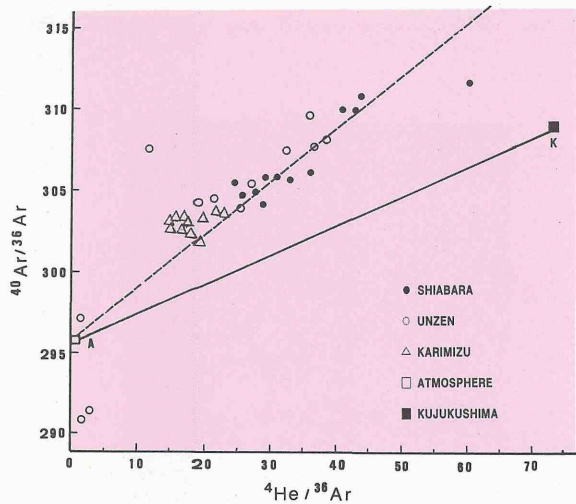


Fig. 12-3. A three-isotope plot of $^{40}\text{Ar}/^{36}\text{Ar}$ vs $^4\text{He}/^{36}\text{Ar}$. A correlation line (dashed line) is defined by data for Shimabara Spa except for S-4 and air-saturated water. The line can be regarded as a mixing line between the atmospheric and magmatic components. Data for the Kujukushima fumarolic gas are plotted far from this line. This suggests two possibilities of elemental fractionation and different sources of magmatic gases. See text.

Conclusions

The isotopic ratios of He, Ne and Ar, and the abundance of five stable noble gases have been determined for gas samples collected at the summit vent, Shimabara Spa, Unzen Spa and Karimizu Spa. The He and Ar isotopic ratios and the abundance ratios have decreased for bubble gases collected at Shimabara after the lava eruption of Unzen Volcano. This suggests that the noble gas composition at Shimabara Spa is one of indicators for the magmatic activity of Unzen Volcano, although there remain some questions to be solved.