A Study on Sulfur Isotope Ratios in Hakone Volcanic Gas

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Hakone Volcano is an active volcano located in western part of Kanagawa Prefecture. Earthquakes swarms began at the end of April 2015, and a small phreatic eruption occurred on June 29. After that, earthquake swarms occurred again in May 2019. Even now, volcanic gas is actively discharged from fumaroles in Owakudani geothermal area. Volcanic gases contain sulfur components such as SO_2 and H_2S, and the sulfur isotope ratio of the sulfur components is one of the useful parameters to track the behavior and processes of volcanic gas components and to estimate the equilibrium temperature. In this study, we measure the sulfur isotope ratios of SO_2 and H_2S in volcanic gases, and discuss the behavior of volcanic fluids and equilibrium temperatures inside Hakone volcano.
Volcanic gases were collected at the Owakudani fumarole (15-2) on a monthly basis from January 2019 to May 2020. This fumarole was formed during a phreatic eruption in 2015. Titanium pipe was inserted into the fumarole to collect volcanic gas, and the interstice between the tube and the outlet of fumarole was closed with soil to prevent air contamination. The titanium tube from the fumarole was connected to a rubber tube, and the volcanic gas was collected by suction into an absorption tube containing a mixed solution of potassium iodate and potassium iodide, or into a vacuum container containing potassium hydroxide. SO_2 and H_2S in the collected volcanic gases were oxidized to $BaSO_4$ precipitation, and the sulfur isotope ratios were determined using a continuous flow stable isotope ratio mass spectrometer.
The measured sulfur isotope ratios of SO_2 ($\delta^{34}S_{CDT}$) were +13.8‰ to +16.2‰, H_2S ranged from -4.5‰ to -3.3‰, and the total sulfur component (ΣS) ranged from +2.3‰ to +8.7‰. Looking at the $\delta^{34}S$ values time variation, the $\delta^{34}S$ of SO_2 peaked around May 2019, when the number of earthquakes increased, and then decreased by 2.4‰, followed by a gradual upward trend. In contrast, the $\delta^{34}S$ of H_2S did not change much. The $\delta^{34}S$ of ΣS increased by 6.4‰ after peaking in August 2019, and then showed a decreasing trend. This suggests that the change in the number of earthquakes and the $\delta^{34}S$ values of SO_2 and ΣS are correlated during the active period of Hakone volcano.
We compared the chemical equilibrium temperature derived from the volcanic gas compositional fractions collected during this study (Ohba et al., 2010) with the isotopic equilibrium temperature determined from sulfur isotope ratios (Richet et al., 1977). The chemical equilibrium temperatures ranged from 276℃ to 513℃, while the isotopic equilibrium temperatures were generally lower, ranging from 205℃ to 237℃. The chemical equilibrium temperature may reflect conditions at high temperatures at depth, while the isotope equilibrium temperature may reflect isotope exchange at relatively low temperatures at shallow depths. Regarding the chemical equilibrium temperature, May 2019, October 2019, and March 2020 showed a large increase of more than 160˚C compared to their respective previous months. In these three large increases in the chemical equilibrium temperature, there is a sharp increase in H_2/H_2O. The magmatic fluid dominated region is considered to have been infiltrated by the surrounding reductive, H_2-rich fluid. On the other hand, the isotope equilibrium temperature peaked around May 2019, increased by about 30˚C compared to February 2019, and has not changed significantly since then. This suggests that the temperature inside the volcano increased during the active phase of Hakone volcano in 2019. The environment inside the volcano, where isotope exchange is occurring, may remain relatively hot after the active period until May 2020.

References

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