Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.37536

On-site analysis of volcanic gas $^{3}He/^{4}He$ at Kusatsu-Shirane Volcano using a portable mass spectrometer

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The helium isotope ratio ($^{3}He/^{4}He$) is a useful indicator for volcanic monitoring. During periods of increased volcanic activity, the contribution of mantle-derived components to volcanic gases increases, resulting in higher $^{3}He/^{4}He$ ratios because mantle helium has a higher $^{3}He/^{4}He$ than atmospheric or crustal helium. Conventional noble gas analysis relies on large magnetic-sector mass spectrometers and requires repeated field sampling followed by laboratory analysis, which has limited the frequency of data acquisition. To address this limitation, we are developing a portable instrument capable of in situ measurements. Discrimination of ^{3}He from HD^{+}, the latter being ubiquitous in a vacuum at a level that interferes with ^{3}He detection, requires a mass resolution ($M/\Delta M$) of ~510. Conventional time-of-flight (TOF-MS) and quadrupole mass spectrometers cannot simultaneously achieve sufficient mass resolution and compactness. We therefore employed the portable "infiTOF" mass spectrometer (KANOMAX JAPAN INC.), derived from the multi-turn time-of-flight mass spectrometer MULTUM-S II, which combines portability with high mass resolution sufficient to resolve $^{3}He^{+}$ and HD^{+} (Toyoda et al., J. Mass Spectrom., 2003; Shimma et al., Anal. Chem., 2010). Noble gas analysis typically requires an ultra-high-vacuum preparation line to remove active gases and separate noble gas species. In this study, we aimed to reduce the size and weight of the overall system by using a heated quartz tube to selectively collect helium from sample gases (Bajo et al., Mass Spectrom., 2012). Field tests were conducted near Kusatsu-Shirane volcano, where long-term $^{3}He/^{4}He$ datasets for several fumaroles are available (Obase et al., Sci. Rep., 2022). The major technical challenges were (1) removal of water vapor, which constitutes more than 95% of fumarolic gases in most subduction zones, including Japan, and (2) continuous pumping of weak fumarolic emissions without atmospheric contamination to supply sample gas to the quartz tube for ~1 h, which is required to accumulate sufficient helium for precise $^{3}He/^{4}He$ analysis. Nevertheless, magmatic helium signatures were successfully identified in several samples, demonstrating the capability of the system for volcanic monitoring.