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Changes in dissolved halogen and sulfur species concentrations over the past 60 years in Yugama, an active crater lake of Kusatsu-Shirane Volcano, Japan

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Yugama, an active crater lake of Kusatsu-Shirane Volcano, serves as a window into the volcano's hydrothermal system. Changes in its lake water composition have been monitored over the long term, providing crucial data for material science-based assessment of volcanic activity.

In this study, we reanalyzed dissolved halogen (fluoride, chloride, bromide, and iodide ions) and dissolved sulfur species (sulfate and polythionate ions) concentrations in Yugama lake water samples stored in our laboratory, collected from 1966 to 2025, using identical analytical methods, revealing long-term temporal changes in anion composition.

These analyses were performed without ion chromatography to ensure analytical reliability. Instead, ion-selective electrode measurements under the coexistence of masking agents for metal ions (for fluoride ions), ICP-OES measurements without chemical pretreatment (for chloride, bromide, iodide ions), and ICP-OES measurements of samples pretreated with anion exchange (for sulfate, polythionate ions) were employed.

By comparing the 60-year variations in anion composition in the Yugama water obtained with the known volcanic activity history of Kusatsu-Shirane Volcano, we evaluated the trend in anion composition fluctuations corresponding to the rise and fall of volcanic activity. First, it was clearly demonstrated that during periods of heightened volcanic activity, polythionate ions are present in significant concentrations. In contrast, during quiescent periods, polythionate disappears, leaving only sulfate ions as the dissolved sulfur species. These changes in the sulfur species composition of Yugama water generally correspond to changes in the chemical composition of gases from ground surface fumaroles around the Yugama, and are thought to reflect fluctuations in the SO_2/H_2S concentration ratio of the gases from subaqueous fumaroles in Yugama. Second, while F, Cl, Br, and I generally show synchronous concentration fluctuations, examining variations in their concentration ratios reveals distinct behaviors. For example, the fluctuations in the concentration ratios of F, Br, and I relative to Cl do not align either in the short or long term. However, at present, no clear answer has been obtained about the factors causing these fluctuations. The differences in halogen behavior in the Yugama water may serve as a useful tool for gaining a deeper understanding of the hydrothermal system of Kusatsu-Shirane volcano.