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Contribution of mantle-derived fluids to the Hishikari gold veins based on ammonium isotope signature

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Ammonium minerals, including buddingtonite (ammonium feldspar) and tobelite (ammonium mica), have been reported from precious-metal epithermal deposits around the world. These minerals have been mainly identified using hand-held short-wave infrared spectrometers (SWIR). Our study identified ammonium-bearing minerals in southern Kyushu and compared the XRD and SWIR results, and measured the ammonium contents and nitrogen isotope compositions. Samples included Au-rich veins and adjacent wallrocks, altered andesites and Shimanto Group basement shales and sandstones from the Hishikari, Fuke, Kago and Kasuga mines as well as the Tobaru prospect. The ammonium contents vary from 193 to 3770 ppm. Concentrations in altered rocks positively correlate with K and Rb in individual districts, confirming substitution of K^+ by NH^{4+}. SEM-EDS indicated that fine-grained illite is the main host of ammonium. Altered sedimentary and volcanic rocks show high $\delta^{15}N$ values, ranging from +16.5 to -2.6 ‰, with an average of +5.5±5.5 ‰ (n= 15). This indicates that sediments and meta-sediments are the major source of ammonium during alteration, since crustal rocks, particularly metamorphosed rocks, have high $\delta^{15}N$ values (typically >+5 ‰). In the Hishikari area, altered rocks also have high ammonium contents and positive $\delta^{15}N$ values, as well as high As and Sb concentrations. By contrast, auriferous adularia-quartz veins at Hishikari contain relatively low ammonium amounts, from 193 to 437 ppm, possibly due to loss of volatile ammonium during boiling of auriferous fluids. Bladed carbonate and colloform quartz provide evidence for boiling during vein formation. The $\delta^{15}N$ values of these gold veins including Kago veins are low, from +4.7 to -4.9 ‰, with an average of +0.45 ±3.8 ‰ (n=8). There is a broad negative correlation between $\delta^{15}N$ values and Au contents for the vein samples, with higher Au correlating with lower ^{15}N values. The $\delta^{15}N$ values of asthenospheric mantle fluids are low, < –6 ‰. Since it is known that boiling does not significantly modify nitrogen isotope compositions, the light, even negative, $\delta^{15}N$ values for the gold veins from Hishikari and Kago suggest a contribution of mantle fluids to the Au-bearing quartz-adularia veins.