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Tracing the extent of fluid circulation in subduction zone forearcs using lithium isotopes

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Despite the recognition that fluids play an important role in subduction zone processes, the extent of fluid circulation and fluid-rock interactions within subduction and accretionary complexes is still not fully understood. Here, we examined Li elemental and isotopic systematics in fluid inclusions trapped within hydrothermal quartz veins in metasedimentary rocks from three paleo-accretionary complexes (Kodiak complex, Alaska; Shimanto Belt, Japan; Western Alps), which are contemporaneous with the burial and metamorphism at temperatures ranging from 250 to 400°C. To provide a fuller understanding, we investigated (i) fluid inclusions, (ii) host quartz, and (iii) wall-rocks of syn-subduction veins.

The $\delta^7\text{Li}$ of fluid inclusion leachates range from -1.5‰ to $+17.1\text{‰}$ and are variable among three localities. Two important processes control the $^7\text{Li}/^6\text{Li}$ ratios of fluids from inclusions: (i) Li release/uptake from the host rock, and (ii) the reactive volume of the rock. Higher $\delta^7\text{Li}$ values of fluids in Kodiak ($+8.1\text{‰}$ to $+17.07\text{‰}$) are interpreted as a result of closed-system behavior, with a small reactive volume of metasediments. Lithium has not been lost to the fluid, where ^6Li is dominantly preserved in metamorphic chlorite and illite. In closed-system samples from the Western Alps, the fluids are buffered by the host rock, causing a shift in $\delta^7\text{Li}$ values of pore fluids (from -1.5‰ to $+9.5\text{‰}$) towards the values of the protolith. Conversely to the samples from Kodiak, the reactive volume of rock is significantly greater, resulting in a complete fluid-rock equilibration. Equally low $\delta^7\text{Li}$ values of pore fluids in Shimanto ($+2.53\text{‰}$ to $+10.39\text{‰}$) is attributed to the large flow of externally derived fluids and interpreted to result by Li leaching from illite and chlorite.

The $\delta^7\text{Li}$ values of quartz are globally higher than those of paired leachates ($+10.93\text{‰}$ and $+22.61\text{‰}$) without temperature-dependent isotopic fractionation between quartz and fluid. This is explained by either (i) a significant drop in pore fluid pressure which, in turn, facilitates rapid

crystallization of quartz, or (ii) post-entrapment re-equilibration between fluid inclusions and the host quartz.

By comparing the metamorphic fluids in the present study with seawater or pore water from deep sea sediments, elevated Li concentrations in leachates (up to 24 ppm) combined with relatively low $\delta^7\text{Li}$ values indicate that Li is progressively leached from sediments during burial, and that the $\delta^7\text{Li}$ value of fluids is consequently shifted towards the signature of the protolith. Similarities in Li concentrations and $\delta^7\text{Li}$ values between leachates and fluids expelled through mud volcanoes in modern examples of subduction zone forearcs further confirms the origin of mud volcano fluids dominantly from subducted sediments. Such similarities imply that fluid circulation across permeable zones may reach at least a 20 km-scale in the forearc region. This study further demonstrates the relevance of Li elemental and isotope systematics to efficiently trace fluids across large distances within subduction zone forearcs.