



Lithium isotopes in terrestrial carbonates as a proxy of naturally occurring arsenic

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Arsenic (As) is one of the most serious inorganic contaminants in drinking water and its occurrence and re-distribution behaviors have been discussed for decades. In the current study, we investigated trace elements compositions of terrestrial carbonates, two tufas and a stalagmite, collected in southwestern Japan. The ambient water of study localities showed As concentrations lower than the drinking criteria of WHO (10 ppb). We found clear positive correlations between As and lithium (Li) concentrations in terrestrial carbonates ($R^2=0.43-0.60$). Since Li isotopic fractionation is understood as an association with chemical weathering and adsorption onto clay minerals, Li isotopic signatures can be applied for further understanding of As releasing processes in groundwater during chemical weathering. We reconstructed the seasonal variation of As and Li concentrations and Li isotopic variations in karstwater from annually laminated tufa sediments. On the other hand, a stalagmite developed during 4000–18000 years B.P. would give geohistorical behaviors of As driven by climate change. To ensure the assumed chemical processes, we also demonstrated a thermodynamic modeling with a hypothetical closed system in karst water. The Li and As concentrations of the stalagmite and tufas were analyzed by quadrupole ICP-MS (Q-ICP-MS, Elan DRCII) at Kochi Core Center and High Precision ICP-MS (Thermo Finnigan ELEMENT2) combined to laser ablation system at National Cheng Kung University, respectively. Li isotopic ratios ($7/6$) were analyzed by MC-ICP-MS (Thermo Finnigan NEPTUNE) at National Cheng Kung University. This study gives a new insight into naturally occurring As systematics.