In situ Sr isotope analysis of methane-seep carbonates: insight into ancient subseafloor fluid circulation

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Along continental margins, tectonic activities trigger subseafloor migration and seepage of geofluids enriched in methane, which sustain chemosynthesis-based biological communities on the seafloor. Depending on their source and migration pathway, seeping fluids have various elemental and isotopic signatures that provide insights into subseafloor fluid–rock interactions and material cycling. At the methane seeps, precipitation of carbonate is induced by microbial oxidation of methane and resultant production of HCO_3^- ions. The methane-seep carbonates record elemental and isotopic signals of seeping fluids in the geological past, and several studies have aimed to decode the origin and migration pathways of ancient seeping fluids using geochemical signatures of the carbonates (e.g., Jakubowicz et al., 2015, 2019).

Strontium isotope ratio ($^{87}Sr/^{86}Sr$) is one of the common tracers of fluid circulation and fluid–rock interactions, and has been applied to seep carbonates (Peckmann et al., 2001; Joseph et al., 2012; Jakubowicz et al., 2019). Methane-seep carbonates are often characterized by complex textures composed mainly of clay- or detritus-rich matrix and detritus-free void cements, the latter of which could archive Sr isotope signals of subsurface fluids distinct from near-seafloor sediments. However, millimeter- or smaller-scale cement phases are difficult to sample using the conventional microdrills without contamination from the matrix. In this study, we applied a high spatial-resolution Sr isotope analysis to cement phases of seep carbonates using laser ablation multiple collector inductively coupled plasma mass spectrometry (LA-MC-ICP-MS), in order to detect “pure” isotope signals of ancient seeping fluids. We measured the Sr isotope ratios in isopachous cements of fossil seep carbonates collected from the Hollard Mound (Middle Devonian, Morocco) and Baška (Lower Cretaceous, Czech Carpathians). Results obtained by LA-MC-ICP-MS were consistent with the isotope ratios measured previously by thermal ionization mass spectrometry coupled with microdrill sampling technique (Jakubowicz et al., 2019). We also tested the method using radial calcite cements of Upper Cretaceous seep carbonates in Hokkaido that are hosted by forearc basin sediments (Yezo Group). The Yezo Group is underlain by Jurassic to Lower Cretaceous mafic basement rocks and input of deep-sourced fluids from the basement rocks could be recorded as ^{87}Sr-depleted signals in the carbonates. Pilot results from one of the Hokkaido seep carbonates, Gakkonosawa, yielded Sr isotope ratios less-radiogenic than those for the coeval seawater, implying deep-fluid input. Exploring the ^{87}Sr-depleted signals at other Cretaceous seeps in Hokkaido would provide more insights into the deep subseafloor fluid circulation in the Cretaceous subduction zone at the eastern margin of the Eurasian Continent.

Keywords: Carbonate, Fluid flow, LA-MC-ICP-MS, Methane seep, Strontium isotopes

