Fluid origin and migration process at the Cretaceous subduction zone of Hokkaido, northern Japan: constraints from Sr and Nd isotopes and U–Pb age of methane-seep carbonates

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Methane seeps are seafloor discharges of geofluids which contain hydrocarbons and various elements mobilized by subsurface fluid–rock interactions. The geofluids play an important role not only in energy supply for chemosynthesis-based life, but also in generation of fuel resources, tectonic activity, and material cycling in the subsurface. The evolution of chemosynthesis-based ecosystems at seeps could have been affected by events driving secular changes in the fluid discharge and the composition of seeping fluids, rather than major extinctions and perturbations of photosynthesis-based ecosystems (Kiel and Peckmann, 2019). Geochemical signatures of seeping fluids provide valuable insights into their origin and composition. However, geochemistry and origin of seeping fluids are not well-known for methane seeps of the ancient Earth. Furthermore, the exact age of fluid discharge is often insufficiently constrained, which has hampered our understanding of events driving the fluid discharge in the geological past.

Here, we present a comprehensive approach to constrain the origin of seeping fluid at Cretaceous methane seeps of Hokkaido, northern Japan, using Sr and Nd isotope signatures of seep carbonates. We also demonstrate results of U–Pb dating of the seep carbonates to directly determine their formation age and place the fluid discharge in the context of tectonic history of this area. During the Cretaceous at the eastern margin of the Eurasian continent, thick marine sedimentary successions were deposited in the Yezo forearc basin. These sediments, called the Yezo Group, are now exposed on land in Hokkaido, northern Japan, and host numerous carbonate bodies formed via anaerobic oxidation of methane at methane seeps. The Cretaceous seep carbonates of Hokkaido yield various animal fossils that have provided key implications on the evolutionary history of chemosynthesis-based ecosystems (e.g., Kaim et al., 2008; Kiel et al., 2008; Jenkins et al., 2018). Using laser ablation-multiple collector-inductively coupled plasma-mass spectrometry (LA-MC-ICP-MS), we performed *in situ* Sr isotope analyses of the seep carbonates collected from 5 sites: Gakkonosawa, Omagari, Yasukawa, Kanajirisawa, and Utagoesawa. The $^{87}Sr/^{86}Sr$ ratios measured in sediment-free calcite cements were similar to, or slightly lower than, those of coeval seawater. The lower values indicate input of a ^{87}Sr-depleted, deep fluid originating from mafic basement rocks underlying the Yezo Group, which correspond to a trapped Jurassic oceanic crust (Kawamura, 2004). We would further test this hypothesis using Nd isotope analysis of the carbonates. We also determined the U–Pb age of the cements using LA-MC-ICP-MS and an international calcite reference material. Although the determined ages have large errors, the ages overlapped at the Albian for the Kanajirisawa and Utagoesawa carbonates and at the Campanian for the Gakkonosawa, Omagari, and Yasukawa carbonates. This result indicates at least two pulses of fluid discharge, which probably corresponded to the uplift or subsequent subsidence of accretionary prism in the Albian and episodic deformation of the forearc basin in the Campanian (Ueda, 2005; Tamaki et al., 2009).

Keywords: Carbonate, Fluid flow, Methane seep, Neodymium isotopes, Strontium isotopes, Uranium–lead dating

