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Constraining silica diagenesis in methane-seep deposits

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Silicified fossils and silicified early diagenetic carbonate minerals as well as authigenic silica phases are common in ancient seep limestones. Silicification of calcareous fossils facilitates the preservation of even fine details and is therefore of great interest to paleontologists, permitting a reliable taxonomic identification of the chemosynthesis-based taxa that lived at ancient hydrocarbon seeps. Four methane-seep limestones of Paleozoic, Mesozoic, and Cenozoic age with abundant silica phases are compared in this study; one, an Eocene seep deposit on the north shore of the Columbia River at Knappton, western Washington State, USA, is described for the first time. Its lithology and fabrics, negative δ13Ccarbonate values as low as $-27.6‰$ and 13C-depleted biomarkers of archaea involved in the anaerobic oxidation of methane (AOM) reveal that the carbonate rock formed at a methane seep. The background sediments of the studied Phanerozoic seep limestones contain abundant siliceous microfossils, radiolarian tests in case of the Late Carboniferous Dwyka Group deposits from Namibia and the Late Triassic Graylock Butte deposits from eastern Oregon (USA), diatom frustules in case of the Eocene Knappton limestone and an Oligocene seep deposit from the Lincoln Creek Formation (western Washington State, USA). These microfossils are regarded as the source of dissolved silica, causing silicification and silica precipitation. All seep limestones used in this study are characterized by very similar paragenetic sequences. Silicified fossils include brachiopods and worm tubes, silica cements include microquartz, fibrous microcrystalline silica, and megaquartz. The silica cements formed after the AOM-derived cements ceased to precipitate but before equant calcite spar formed. Numerical experiments using the computer code PHREEQC were conducted to test the hypothesis that (1) AOM increases the pH of pore waters and that (2) this pH increase subsequently mobilizes biogenic silica, (3) followed by the re-precipitation of the dissolved silica in the periphery of the AOM hotspot. The experiments revealed that degassing of carbon dioxide, resulting from AOM-driven carbonate precipitation, is a key factor that has the potential to significantly increase the local pH of pore waters. The results indicate that carbon dioxide degassing exerts an even stronger control on the local pH and silica dissolution than the rate of AOM alone. Numerical experiments demonstrate that AOM in combination with degassing of carbon dioxide is an effective trigger for silica dissolution, allowing for silica re-precipitation at some distance from the AOM hotspot. Our experiments provide a conceptual model for the mechanics of silicification and silica precipitation at seeps.