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Authigenic carbonates record changing redox conditions at cold seeps from Alaminos Canyon, northern Gulf of Mexico

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Anaerobic oxidation of methane in anoxic sediments at cold seeps often leads to formation of authigenic carbonates close to the seafloor along continental margins. Recent work, however, indicated that the redox conditions prevailing during seep carbonate precipitation may vary to some degree. In order to shed new light on the extent of this variability, authigenic carbonates from Alaminos Canyon lease block 645 of the northern Gulf of Mexico have been characterized by means of inorganic and organic geochemistry. The carbonates were collected from seep deposits representing various seafloor morphologies, including extensive pavements, mounds, fractured carbonate slabs surrounded by dense bivalve shells, and vestimentiferan tubeworm colonies. The deposits almost entirely consist of aragonite. The $\delta^{18}O$ values of aragonite vary from +2.6 to +5.8‰ V-PDB, suggesting precipitation in slight disequilibrium with the surrounding pore fluids. The $\delta^{13}C$ values of aragonite between –33.9 and –20.4‰ V-PDB agree with variable amounts of carbonate derived from oxidation of thermogenic methane and crude oil. Methane was primarily oxidized in an anaerobic process as revealed by the presence of ^{13}C-depleted molecular fossils of methane-oxidizing archaea ($\delta^{13}C$ values as low as -118%¸) and sulfate-reducing bacteria ($\delta^{13}C$ values as low as -97%¸), the syntrophic partners in the anaerobic oxidation of methane. The observed inventories of molecular fossils in the authigenic carbonates mirror those of known consortia of anaerobic methane oxidizing archaea (ANME) and sulfate-reducing bacteria, namely the ANME-2/*Desulfosarcina*/*Desulfococcus* (*DSS)* and ANME-3/*Desulfobulbus (DBB)* consortia. In contrast, the same carbonates exhibit shale-normalized rare earth elements patterns that all display real negative Ce anomalies (Ce/Ce* $<$ 0.78), suggesting that precipitation proceeded at least partially under oxic conditions. The episodic occurrence of oxic conditions is confirmed by the presence of molecular fossils of aerobic methanotrophic bacteria, including 4α-methylcholesta-8(14),24-dien-3β-ol, and two bacteriohopanepolyols, aminotetrol and aminotriol. The $\delta^{13}C$ values of the biomarkers of aerobic methanotrophs are as low as –58‰ and are consequently less ^{13}C-depleted than the molecular fossils of the prokaryotes performing anaerobic oxidation of methane; a pattern in accord with culture experiments. Overall, our results suggest that redox conditions at cold seeps are variable. This variability probably reflects changes in seepage flux. The combination of an inorganic and an organic geochemical approach used here is promising to better assess the variability and diversity of past fluid and gas expulsion at seeps.