

Rising bottom-water temperatures induced methane release during the middle Holocene in the Okinawa Trough, East China Sea

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In this study, a piston core was collected from a seep-impacted area at a water depth of approximately 1000 m from the Okinawa Trough, East China Sea, and was studied using comprehensive analyses of carbon and oxygen stable isotopes, lipid biomarkers, and major and trace element geochemistry. Extremely low $\delta^{13}\text{C}$ values of total inorganic carbon and organic carbon, increased Methane Index, and molybdenum (Mo) and uranium (U) enrichments at depths of 225–255 cm below the sea-floor (bsf) and 75–142.5 cm bsf were found. Combined with published pore water data, fossil and current sulfate-methane transitions (SMTs) were identified. Mass balance equations were applied to estimate the fraction and content of authigenic carbonates at the two SMTs and their corresponding $\delta^{18}\text{O}$ values. The calculations revealed that the estimated $\delta^{18}\text{O}$ values of authigenic carbonates for the fossil SMT (from 4.9‰ to 5.8‰) and the current SMT (from -2.1‰ to 4.4‰) were higher and lower than the theoretical equilibrium $\delta^{18}\text{O}$ value (4.8‰), respectively. These results suggest that fossil seepage is possibly induced by the dissociation of gas hydrates. The fluids of the current seepage may be derived from the equilibrium between the ambient seawater and the fluids of gas hydrate dissociation. Based on the accumulation of authigenic carbonate in sediments, previously published pore-water calcium and magnesium fluxes, and foraminifera ^{14}C dating, the fossil methane seepage was confirmed to have occurred during the period 8.2 to 4.5 ka B.P., with its SMT depth less than 105 cm bsf, while the present-day methane seepage with a low fluid intensity was estimated to have started before 1.0 ka and formed its SMT at approximately 250 cm bsf. The timing of this fossil methane seepage suggests that the rising temperature of the North Pacific Intermediate Water during the early and middle Holocene mainly controlled the methane release in the Okinawa Trough, whereas the ongoing methane emission was likely induced by decreased pressure driven by back-arc extension. The geochemical data demonstrate past