Isotope fractionation and mixing in methane plumes from the Logatchev Hydrothermal Field

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As methane is consumed in the deep sea, its \(^{13}C/^{12}C\) ratio progressively increases due to kinetic isotope fractionation. Many submarine hydrothermal vents emit methane with carbon isotope ratios that are higher than those of background methane in the surrounding ocean. Since the latter exists at low concentrations, mixing of background methane with vent fluid tends to decrease the ¹³C/¹²C ratio as concentration decreases, opposite to the trend produced by consumption. We investigated CH₄ concentration and ¹³C/¹²C together with ³He in plumes from the Logatchev hydrothermal field (LHF), which generates methane with δ^{13} C \approx -13‰ by serpentinization of ultra-mafic rock. The measured methane and δ^3 He were well correlated at high concentrations, indicating a $CH_4/^3$ He ratio of 1×10^8 in the vent fluids. These tracer distributions were also simulated with an advectiondiffusion model in which methane consumption only occurs above a certain threshold concentration. We utilized δ^3 He to calculate the methane remaining in solution after oxidation, f, and the deviation of δ^{13} C from the value expected from mixing alone, $\Delta \delta^{13}$ C. Both in the model and in the data, the entire set of $\Delta \delta^{13}$ C values are not correlated with log f, which is due to continuous oxidation within the plume while mixing with background seawater. A linear relationship, however, is found in the model for methane at concentrations sufficiently above background, and many of the samples with elevated CH₄ north of LHF exhibit a linear trend of $\Delta\delta^{13}$ C versus log f as well. From this trend, the kinetic isotope fractionation factor in the LHF plumes appear to be about 1.015. This value is somewhat higher than found in some other deep-sea studies, but it is lower than found in laboratory incubation experiments.

Cretaceous carbon cycle and climate: The negative carbon isotope spike at the onset of OAE1a in the Early Aptian

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The negative C-isotope spike at the base of OAE1a in the Early Aptian records a CO₂ pulse caused by the Ontong-Java volcanism. Methane gas hydrate dissociation may have contributed to the negative spike. However, it was not the trigger but the result of greenhouse climate conditions.

Lithology, bulk-rock and molecular C-isotopes, Corg- and Ccarb-content and nannoconid abundances have been investigated within the negative spike interval of various Tethyan sections.

Sediment thickness of the studied pelagic sections is extremely variable pointing to a current-controlled sedimentation along deep continental margins of the alpine Tethys.

Lithologies vary from limestones with frequent chert layers to more marly limestones containing prominent marls and black shales. Bulk inorganic and organic as well as molecular C-isotope values feature a negative spike with an amplitude of $\sim 1.4\%$, $\sim 2.5\%$ and up to $\sim 8\%$ respectively, while fractionation between carbonate and organic carbon increases.

Bulk $CaCO_3$ contents and nannoconid abundances drop drastically at the base and within the negative spike interval, which is thought to reflect ocean acidification due to an increase in pCO_2 . The latter can also explain the observed increase in fractionation. It is possible that additional input of isotopically very negative C from the dissociation of methane gas hydrates further amplified the negative spike.