Elucidating ^{13}C-depleted methane generation during anaerobic methanotrophy

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The anaerobic oxidation of methane (AOM) is a key microbial process in the subsurface methane cycle. According to conventional isotope systematics, AOM should leave a residue of methane enriched in ^{13}C, but has been frequently found to be depleted in ^{13}C. This has been interpreted as co-occurrence of methane production and oxidation. However, a recent study gave evidence of ^{13}C-depleted methane generation during AOM based on enzyme-driven isotopic equilibration between methane and DIC, but this effect was rather marginally expressed in methane isotopes. In order to gain a deeper understanding of the enzyme-driven isotopic equilibration during AOM we performed extensive isotope fractionation experiments with thermophilic enrichments of ANME/SRB consortia devoid of methanogens. In incubations with 10 mM sulfate concentrations ^{13}C-methane isotopes became progressively more positive yielding a $\Delta\delta^{13}C_{end\text{-}start}$ of +58‰, while strongly ^{13}C-depleted methane ($\Delta\delta^{13}C_{end\text{-}start}$ = -26‰) was generated during sulfate limitation (< 1 mM). Tracer experiments employing ^{14}C-methane and ^{14}C-DIC at different sulfate concentrations showed only minor increase in the back flux from the DIC into the methane pool, thus excluding carbon isotope equilibration between AOM endmembers. Further isotope labelling experiments suggest the initial activation reaction from methane to methyl-coenzyme M as explanation for the diverging isotope fractionation patterns in AOM. Our results have wide implications on the interpretation of methane isotope profiles and the use of tracers for diverse rate measurements.