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Impact of gas transport on fractionation of carbon stable isotopes related to the microbial oxidation of methane in soils

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Stable isotope (SI) analysis enables the study of in situ methane removal efficiencies in systems where not all flux pools can be directly measured. As the stable methane isotopes ($^{12}CH_4$, $^{13}CH_4$) differ in molecular weight by 6.25 %, their size, steric properties and their diffusion coefficient differ, leading to unequal behaviour at enzyme binding sites and to different transport rates. As a result of this, a fractionation of the isotopes occurs during the process of microbial methane oxidation (Barker and Fritz, 1981) and during diffusive gas transport (Reeburgh et al., 1997; Tyler et al., 1994; De Visscher et al., 2004), resulting in preferential oxidation and faster diffusive transport of the lighter isotope and hence in an enrichment of the heavier isotope in the remaining gas phase.

No fractionation is expected to occur when advection dominates gas flow, so that the fractionation factor $\alpha_{trans\_adv} \cong 1$ (Chanton and Whiting, 1996; Bergamaschi et al., 1998; Chanton, 2005). However, fractionation of stable CH_4 isotopes during gas transport occurs when diffusion governs the transport process. Based on the differences in molecular mass, the binary diffusion coefficient of $^{12}CH_4$ in air in theory exceeds that of $^{13}CH_4$ by a factor of 1.0195, so that α_{trans_diff}= 1.0195 (Marrero and Mason, 1972). This is in the same order of magnitude as fractionation factors found for methane oxidation (α_{ox}), indicating that methane oxidation can be significantly overestimated if calculations based on isotopic fractionation are not corrected for fractionation by diffusion.

Gas diffusivity and gas permeability of soils is closely related to the share of coarse pores > 50 μm (also referred to as air capacity) as these are usually drained and hence available for gas transport. Air capacity in turn is strongly impacted by soil texture and soil compaction. In order to assess the relevance of gas transport on the fractionation of carbon stable isotopes, a column study simulating a landfill cover soil was conducted using a sandy loam compacted to three different levels (75, 85, 95% of the Proctor density; setup in Gebert et al., 2010) and four different flux scenarios: gas transport dominated by advection, with and without methane oxidation; gas transport dominated by diffusion, with and without methane oxidation. In each case, soil gas composition, the $\delta^{13}C$ ratio and the methane oxidation rate were determined in eight depths by means of GC-FID/TCD and GC-IRMS analysis. Results showed that the greatest changes in the $\delta^{13}C$ ratio occur when gas transport is governed by diffusion in combination with methane oxidation (enrichment of ^{13}C by 29 ‰. It was also seen that changes in the $\delta^{13}C$ ratio of up to 20 ‰ can occur through diffusive gas transport only (no oxidation), indicating the necessity to include an appropriate value for α_{trans} when the fraction of methane oxidized (f_{ox}) is calculated from SI data. The negative changes (depletion in ^{13}C) in the ratio between 5 cm depth and the soil surface (0 cm) confirmed phenomena described by Chanton et al. (2008) and are hypothesized to be caused by preferential pathway oxidation, bypass mixing or diffusive fractionation; complicating comparison between gas profile and gas emission data.

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