Effect of methane gas phase dynamics on the coupled methane-sulfur cycles in the subsurface

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Introduction

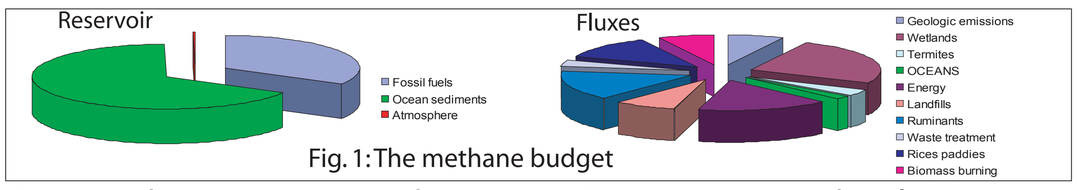

Fig. 1: The methane budget

Ocean sediments represent the greatest CH_4 reservoir on the planet. Nevertheless, their CH_4 contribution is negligible towards the ocean-atmosphere system (fig. 1). This is due to the microbial process of anaerobic oxidation of methane (AOM), which acts as an efficient subsurface CH_4 barrier and thus an important climate regulator. Our scope is to investigate the efficiency and behavior of AOM with respect to the dynamics of free methane gas ($CH_{4(g)}$).

Conceptual model

Particulate organic carbon (POC) degradation coupled to SO_4^{2-} reduction takes place in the upper parts of the sediment. Once SO_4^{2-} is consumed, methanogenesis (MET) begins and generates CH_4 (aq) in the deeper layers of the sediment. When CH_4 (aq) exceeds the in situ solubility, $CH_{4(g)}$ forms. Buoyancy leads to upward gas flow, and dissolution takes place in the overlying undersaturated sediment layers where CH_4 (aq) is consumed by AOM (Fig. 2).

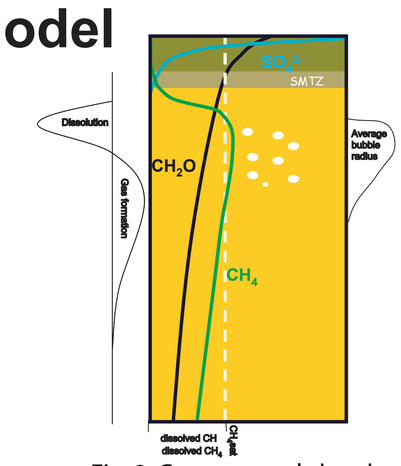

Fig. 2: Concenptual sketch of the processes

Comparison to field data - Profiles

Fig. 3 shows measured (Martens et al., 1998) and simulated CH_4 (aq) and SO_4^{2-} profiles for Eckernförde Bay sediments (Kiel Bight, Germany) at steady state. Supersaturation due to high MET rates leads to CH_4(g) which is transported upwards and dissolves in undersaturated areas near the sulfate-methane transition zone (SMTZ). Gas results are compared to acoustic sounder profiles (fig. 3 left color column) collected by Wever et al., (2006).

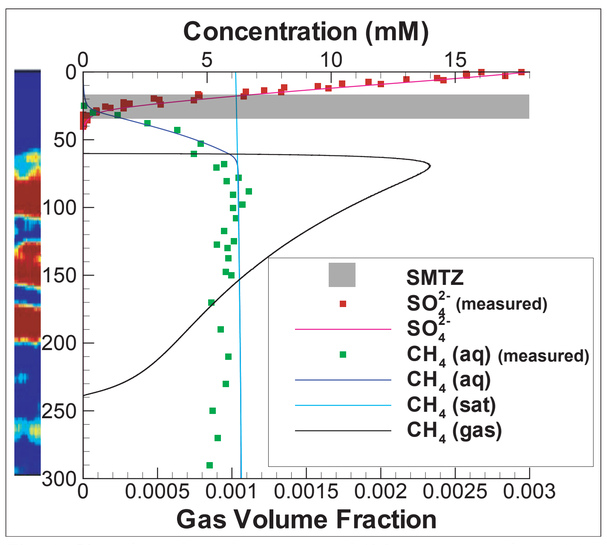

Fig. 3: Simulated and measured (Martens et al., 1998, Wever et al., 2006) geochemical profiles at Eckernförde Bay

Comparison to field data - Rates

CH_4 fluxes towards the SMTZ feed the microbial community performing AOM. In systems with high POC fluxes to the sediment-water interface (SWI), the transport and dissolution of $CH_{4(g)}$ in methane-depleted areas produces a steep CH_4 gradient which maintains high CH_4 diffusive fluxes. Thus, in systems where $CH_{4(g)}$ AOM rates are high and may even overshadow sulfate reduction rates (fig. 4).

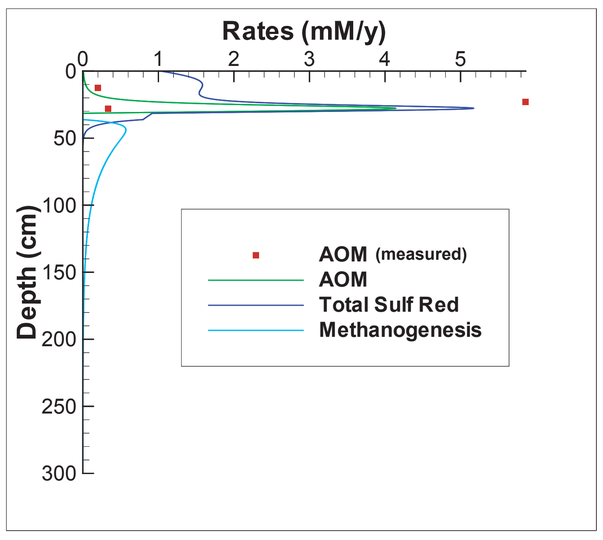

Fig. 4 Simulated and measured (Martens et al., 1999,) geochemical rates at Eckernförde Bay

Gas effects on AOM

Increasing POC fluxes lead to shallower SMTZs which amplify the zone of MET and thus the $CH_{4(g)}$ generated. Upward $CH_{4(g)}$ migration increases the diffusive flux and consequently the AOM intensity (fig. 5). In a system that ignores (implicit - fig. 5) the dynamics of $CH_{4(g)}$, the integrated AOM rate underestimation increases exponentially with higher POC fluxes, demonstrating the importance of $CH_{4(g)}$ feedbacks on AOM (Mogollón et al., 2008).

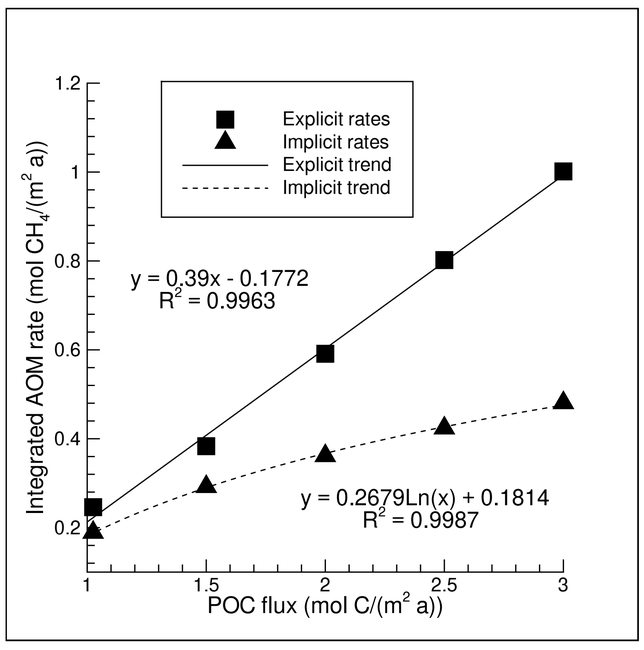

Fig. 5. Integreted AOM vs. POC fluxes

Seasonal variations in $CH_{4(g)}$

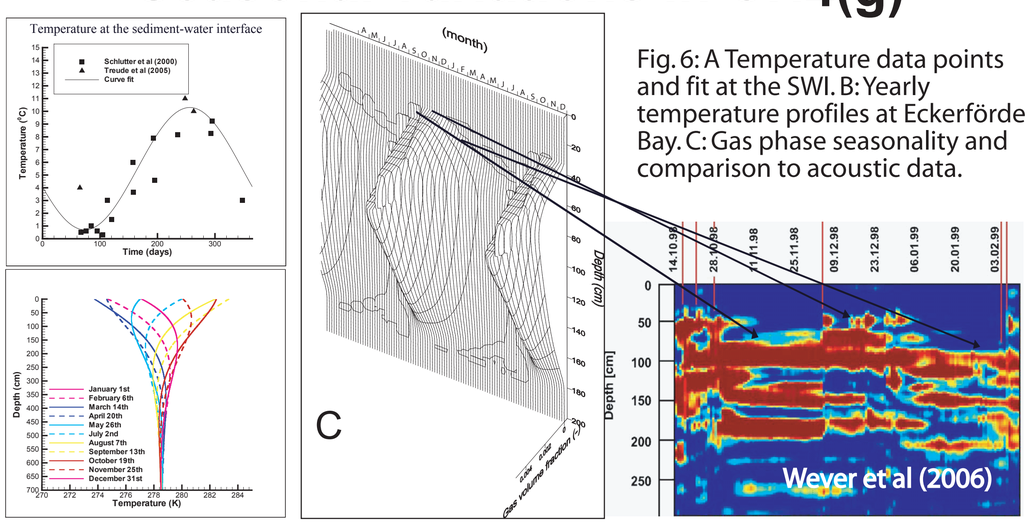

Fig. 6: A Temperature data points and fit at the SWI. B: Yearly temperature profiles at Eckerförde Bay. C: Gas phase seasonality and comparison to acoustic data.

Variations in temperature at the SWI (fig. 6a) lead to heat diffusing into the sediments. The heat capacity of the sediment produces lag times reflected in both the temperature profiles (fig. 6B) and the monthly variations in the methane bubble depth (MBD) (fig. 6C). In the early winter, when the gas is shallowest, the propensity for $CH_{4(g)}$ escape increases.

Predicting CH4 fluxes from MBD

The strong correlation between the MBD and CH_4 fluxes to the SMTZ can lead to predictions of the CH_4 turnover rates (integrated AOM rates) when $CH_{4(g)}$ escape from the sediment is negligible. Dale et al. (2008) developed MBD-CH_4 flux curves for various CH_4 saturations based on acoustic survey profiles (a standard mapping technique) and measured AOM rates. Our preliminary seasonal simulations show the same effect (fig. 7).

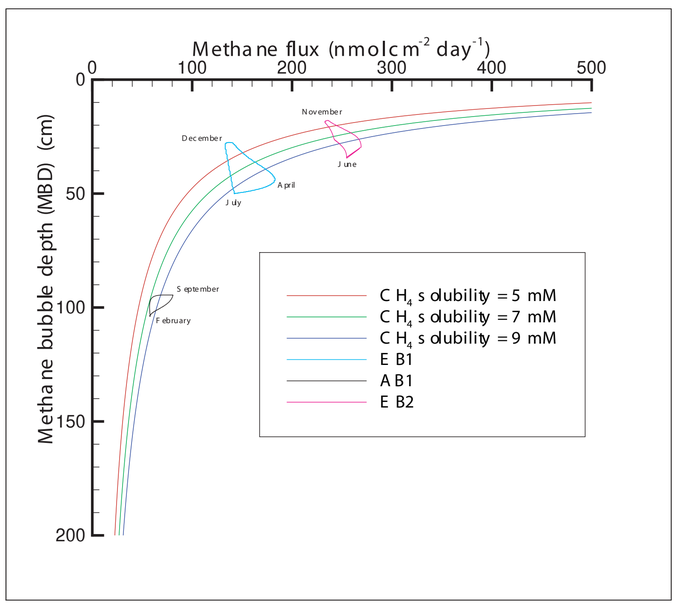

Fig. 7. Methane fluxes as a function of the methane bubble depth

Conclusions

- $CH_{4(g)}$ produced in shallow sediments with high POC content has a greater propensity towards migration than dissolved methane.
- $CH_{4(g)}$ transport effectively delivers CH_4 to the SMTZ and leads to increased AOM rates.
- AOM in Eckernförde Bay is strong enough to consume both dissolved and gaseous methane leading to negligible gas escape into the water column.
- Understanding $CH_{4(g)}$ dynamics can lead to the development of cost-effective tools that can predict CH_4 fluxes and integrated AOM rates.

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