Seasonal controls of methane gas solubility and transport on anaerobic oxidation of methane in shallow water marine sediments

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Introduction

Particulate organic carbon (POC) degradation coupled to SO_4^{2-} reduction takes place in the upper parts of anoxic sediments. Once SO_4^{2-} reaches sub-mM concentrations, methanogenesis (MET) begins. When MET is high, the local CH_4 (aq) may exceed the CH_4 solubility leading to CH_4 (g) formation. Upward gas migration and dissolution in the overlying undersaturated sediment enhance methane consumtion in the presence of SO_4^{2-} through a process known as anaerobic oxidation of methane (AOM). If sufficient, gas may also escape the AOM barrier and leave the sediment. Seasonal variations in the CH_4 solubility can leat to times of preferetial CH_4 (g) production and CH_4 (g) dissolution (Fig 1), which are crucial for determining the methane cycle.

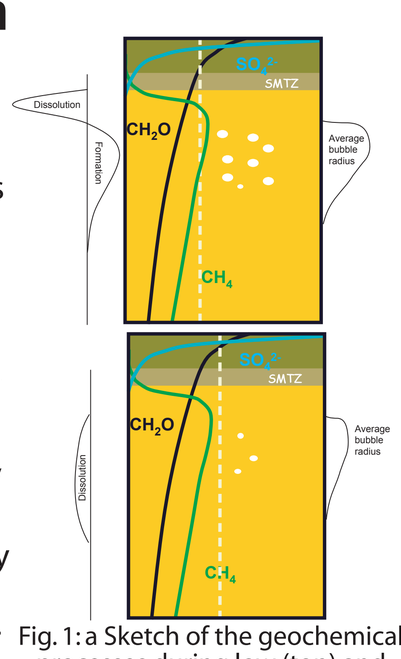


Fig. 1: a Sketch of the geochemical processes during low (top) and high (bottom) CH_4 saturation

Controls on methane solubility

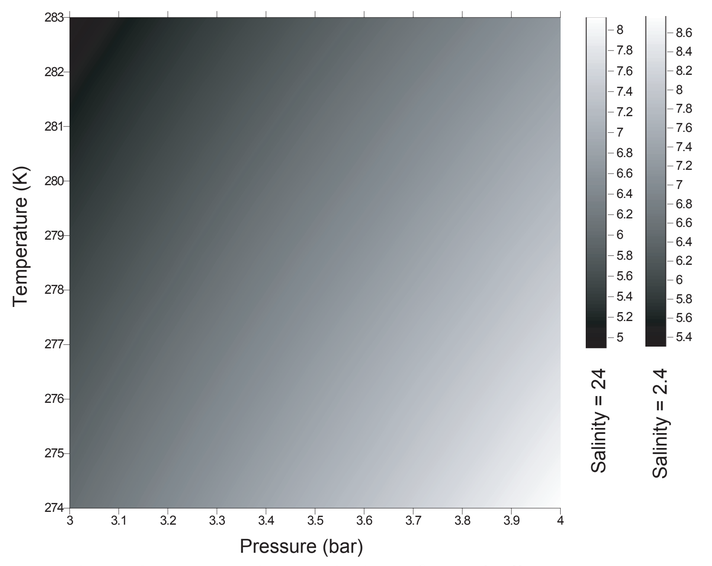


Fig. 2. CH_4 saturation variations for a shallow (< 30 m) marine to brackish sediment

The solubility of free methane gas depends on the local pressure, temperature and salinity. Figure 2 shows a calibrated algorithm that predicts the methane solubility as a function of these parameters for shallow water conditions. These parameters may vary accros time

Steady-state calibration

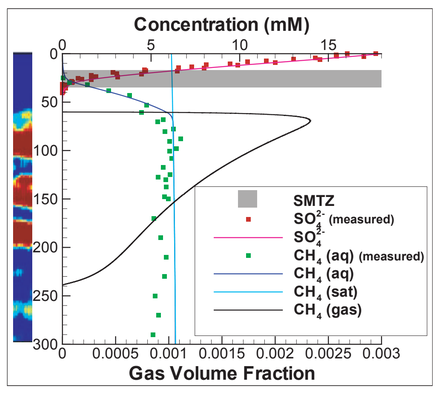


Fig. 3: Simulated and measured (Martens et al., 1998, Wever et al., 2006) geochemical profiles at Eckernförde Bay

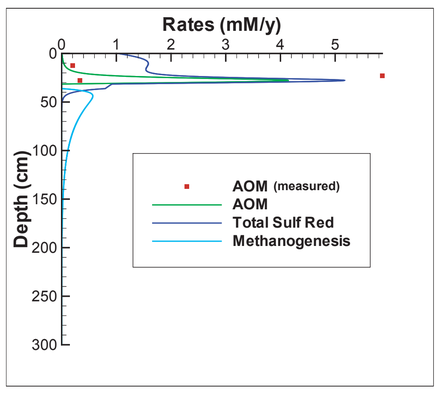


Fig. 4 Simulated and measured (Martens et al., 1999,) geochemical rates at Eckernförde Bay

A 1D, 3-phase model was built to explore the fate of CH_4 (g) in Eckernförde Bay (Mogollón et al., 2009). The model was calibrated with measured concentration and rate profiles from Martens et al (1998, 1999) (Figures 3,4)

Pressure effects on gas inventory

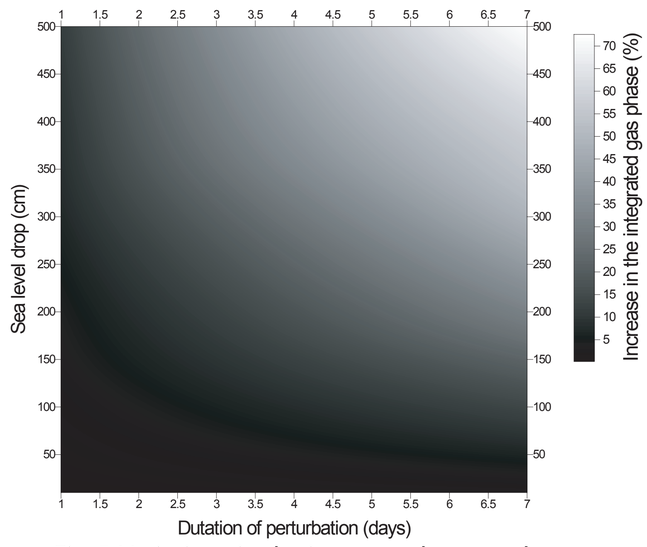
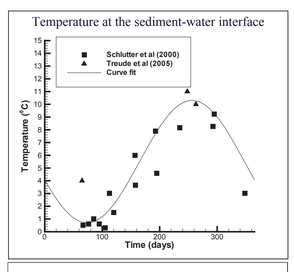
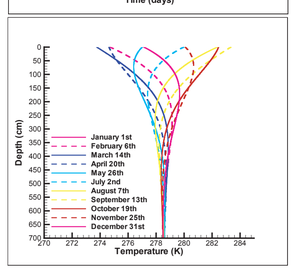


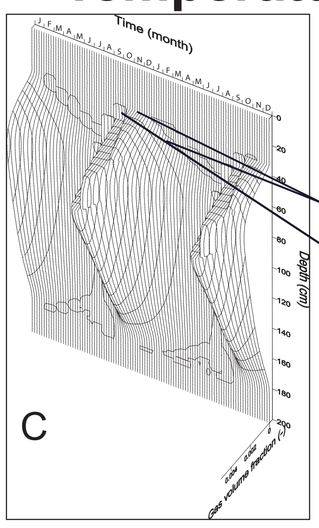
Fig. 5. Variations in the integrated gas pool with respect to the steady-state values (Figs 2,3) due to sustained pressure drops

During pressure drops, the methane solubility will decrease and stimulate gas formation in the porewater. The magnitude of the gas formation and its increase with respect to the steady-state gas pool will depend on both the magnitude and the duration of the pressure drop (Fig 5). For Eckernförde Bay, tidal fluctuations and atmospheric pressure changes are not long and sufficient enough to trigger gas escape.

Temperature







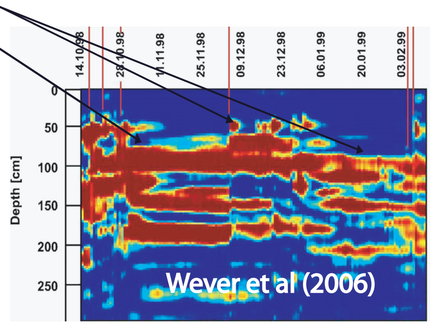


Fig. 6: A Temperature data points and fit at the SWI. B: Yearly temperature profiles at Eckerförde Bay. C: Gas phase seasonality and comparison to acoustic data.

Variations in temperature at the sediment-water interface (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.

Gas effects on AOM

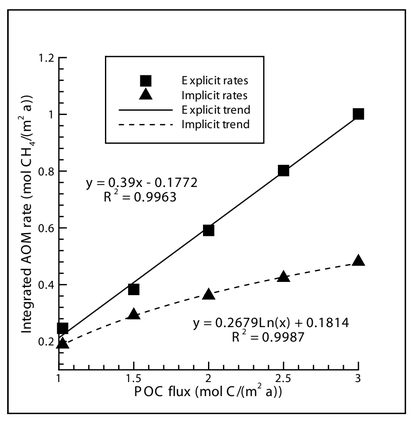


Fig. 7. Integreted AOM vs. POC fluxes

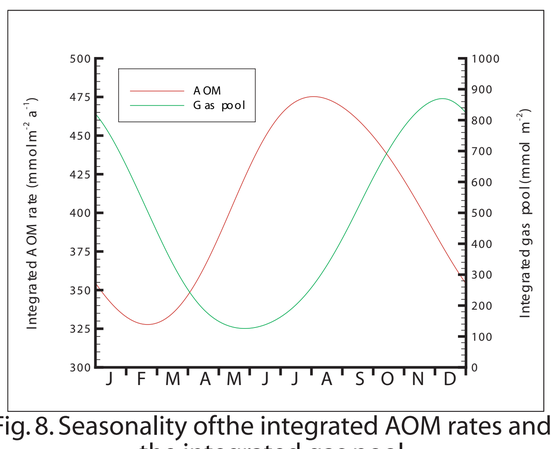


Fig. 8. Seasonality ofthe integrated AOM rates and the integrated gas pool

Increasing POC fluxes increase the SR rates, amplifying the zone of MET and thus the CH_4 (g) generated and AOM intensity. If CH_4 (g) is ignored (implicit - Fig. 7), integrated AOM rates reach a plateau and may be underestimated with respect to simulations that include CH_4 (g) (explicit - Fig. 7). Furthermore, a temperature drop will both decrease in the intensity of microbial activity and increase gas dissolution rates (Fig. 8). These opposing processes for AOM will shift in magnitude for sediments according to the MBD: When shallow, temperature dominates and when deep gas dissolution dominates.

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. 9).

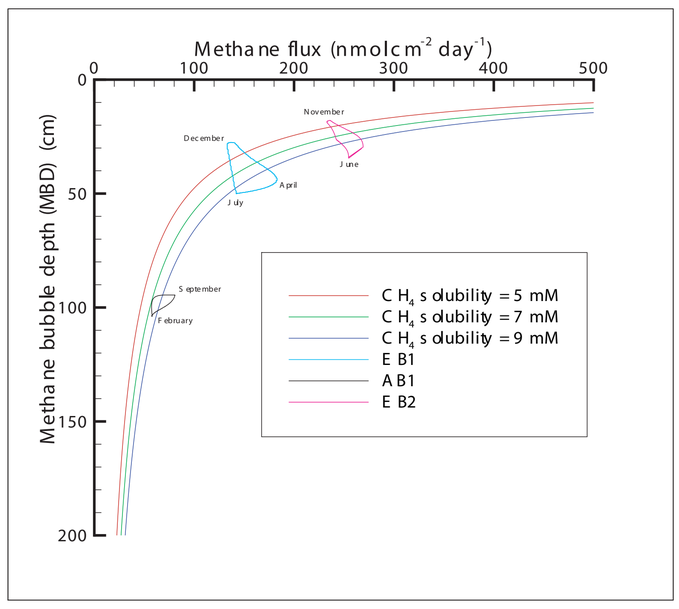


Fig. 9. Methane fluxes as a function of the methane bubble depth

Conclusions

- Seasonal variations in temperature at the SWI lead to pronounced changes in solubility and, consequently the methane bubble depth.
- Pressure drops due to storms and intense wind activity may lead to ample gas formation, but at the daily time scales these are too short to allow gas escape for MBDs > 50 cm
- Seasonal AOM cycles are influenced by the dissolution of CH_4 (g), which sustains the CH_4 (aq) concentrations as methane is consumed in AOM.

Acknowledgements:

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