



Quantification and controlling factors of methane loss and storage within marine sediments along a transect off Peru

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The driving force of biogenic methane formation is the irreversible redox-conversion of organic matter. Our modelling approach incorporates interdependent diagenetic reactions evolving into a diffusive multi-component and multi-phase system by means of thermodynamic equilibrium calculations of species distribution. The modelling tool is the PHREEQC (version 2) computer code (Parkhurst and Appelo, 1999). Reaction kinetics of organic carbon conversion is integrated into the set of equilibrium reactions by defining type and amount of converted organic matter in a certain time step. The model setup describes a growing sediment column. One-dimensional molecular diffusion of aqueous and gaseous species as well as burial of solids and aqueous species are considered. Carbon mass balance calculations along a transect from the shelf to the open ocean off Peru (ODP Leg 112 and 201; D'Hondt et al., 2002; Suess et al., 1988) highlight the influence of redox-conversion of organic matter on methane and diagenetic carbonate formation. After conversion of organic carbon the final carbon pool is dominated by secondary carbonate minerals. Additionally, aqueous CH₄ and methane hydrate form, and modelled dissolved methane concentration profiles match very well measured profiles. In total, all investigated sites obtain a moderate carbon loss, with exception of the open ocean site.

Our model results indicate that at the shelf sites, carbon loss is regulated by the amount of authigenic carbonate formation, neither methane nor CO₂ are significant in carbon mass balance calculations. Significant methane concentrations establish in deeper sediments (Miocene age) of the shelf edge. At the lower slope, two sites (682 and 688) were examined. High organic matter conversion in upper sediment layers at site 682 leads to high CO₂ and methane production close to the sediment surface. Consequently, the diffusive loss of methane and CO₂ into overlying sea water is high and most of the converted organic carbon (92 %) gets lost. On the other hand, very high organic matter conversion in deeper Holocene/Pleistocene sediments of site 688 together causes the formation of final carbon amounts in the sediments that are more than twice as high as at other sites. Most of the converted carbon is stored within the sediments because of high sedimentation rates that prevent gaseous and aqueous species from diffusion into the overlying sea water. Furthermore, methane is stored in the form of hydrates within the sediments.

Carbon mass balance calculations based on our modelling clarify that diffusion controls the carbon cycle, and especially methane accumulation in or release of marine sediments. Crucial for the diffusive fluxes of methane or carbon dioxide out of the sediments are concentration gradients between pore water, sea water, and sedimentation rates. When sedimentation rates are high, the carbon loss from the system is low, whereas low sedimentation rates together with high organic matter conversion lead to high diffusional fluxes of dissolved methane and CO₂ out of sediments.

Following these model results, especially lower slope settings that exhibit very high sedimentation rates due to the input of allochthonous organic-rich sediments from the shelf are most promising for methane accumulation and carbon storage. On the other hand, shelf sediments with lower sedimentation rates but high organic matter conversion close to the sediment surface match a carbon source.

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