

## Methane flux in potential hydrate-bearing sediments offshore southwestern Taiwan

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Methane in interstitial water of hydrate-bearing marine sediments ascends with buoyant fluids and is discharged into seawater, exerting profound impacts on ocean biogeochemistry and greenhouse effects. Quantifying the exact magnitude of methane transport across different geochemical transitions in different geological settings would provide bases to better constrain global methane discharge to seawater and to assess physio-chemical contexts imposed on microbial methane production and consumption and carbon sequestration in marine environments. Using sediments collected from different geological settings offshore southwestern Taiwan through decadal exploration on gas hydrates, this study analyzed gas and aqueous geochemistry and calculated methane fluxes across different compartments. Three geochemical transitions, including sulfate-methane transition zone (SMTZ), shallow sediments, and sediment-seawater interface were specifically focused for the flux calculation. The results combined with previous published data showed that methane fluxes at three interfaces of  $2.71 \times 10^{-3}$  to  $3.52 \times 10^{-1}$ ,  $5.28 \times 10^{-7}$  to  $1.08 \times 10^0$ , and  $1.34 \times 10^{-6}$  to  $3.17 \times 10^0$  mmol m $^{-2}$  d $^{-1}$ , respectively. The ranges of fluxes suggest that more than 90 % of methane originating from depth was consumed by anaerobic methanotrophy at the SMTZ, and further >90% of the remnant methane was removed by aerobic methanotrophy prior to reaching the sediment-seawater interface. Exceptions are sites at cold seeps where the percentage of methane released into seawater can reach more than 80% of methane at depth. Most sites with such high methane fluxes are located at active margin where thrusts and diapirism are well developed. Carbon mass balance method was applied for the calculation of anaerobic oxidation of methane (AOM) and organotrophic sulfate reduction rates at SMTZ. Results indicated that AOM rates were comparable with fluxes deduced from concentration gradients for most sites. At least 60% of sulfate infiltrating from seawater was consumed by AOM. Gas compositions and methane carbon isotopes show microbial gas dominated at passive margin and lower slope of active margin; by contrast, thermogenic gas source was prevalent at upper slope of active margin. In summary, transport of deeply sourced methane in potential hydrate-bearing sediments is strongly controlled by geological structures and microbial processes. For most of sites, anaerobic and aerobic methanotrophy in sediments act as efficient biofiltration for the removal of methane. For sites with strong fluid advection, a great fraction of deeply-sourced methane could escape from anaerobic and aerobic methanotrophy and be discharged into seawater column. The changing mechanisms for gas generation from passive to active margin highlights the interplay between in situ methanogenesis, sediment loading, and connectivity of fluid conduits.