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A BIOGEOCHEMICAL PERSPECTIVE ON SORBED GASEOUS HYDROCARBONS AND ITS VERTICAL DISTRIBUTION IN MARINE SEDIMENTS

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Methane adsorbed to the mineral phase plays an unknown role within biogeochemical and geological processes. The amount of sorbed gas, its chemical bondings, mechanisms and kinetics of sorption processes are poorly constrained. Potential adsorbents for hydrocarbon gases are organic carbon, carbonate and clay minerals, especially 2:1-layered clays.

In laboratory autoclave experiments, we could show that purified smectites (Na-Montmorillonite) can hold up to 92 $\pm$ 2 mmol CH_4 per kg dry mineral, whereas the sorption capacity of kaolinite was calculated to 4 mmol CH_4 kg^{-1}. Changes in stable isotopic compositions of dissolved and sorbed methane were observed in smectite minerals. Weakly-bound gas did not fractionate significantly, whereas strongly-bound methane extracted after a 50-day-period was depleted in ^{13}C by -7 per mil relative to the dissolved gas.

We compared different gas extraction techniques, such as boiling of samples in phosphoric acid, autoclavation, and subjecting samples to a 1-N-NaOH solution for a prolonged period of time, and found that base extraction is far more efficient than the

conventional acid extraction (95 to 99% more methane) and autoclavation (29 to 92% more methane). We suggest that under highly basic conditions silicates dissolve and subsequently delaminate and release the gas trapped in interlayer spaces.

Concentrations of sorbed methane from more than 300 environmental samples ranged within five orders of magnitude from 4.8×10^{-5} to 2.6 mmol L^{-1} wet sediment. In certain environments, significantly more gas is present in a sorbed than dissolved state. In down-core profiles, maximum amounts of sorbed methane do not correlate with solid phases but with indicators for microbial activity, thus indicating a biogeochemical control on the amount of sorbed gas. At specific sites within the Black Sea (Pechori Mound, Georgia, BS-359-AP), higher alkanes such as ethane to hexane were extracted at higher levels than methane. Stable carbon isotopic values of sorbed methane ranged between –78.8 and –48.1 per mil. A general similarity of isotopic values of sorbed and associated dissolved methane suggests a dynamic exchange between both pools. Therefore the source of sorbed CH_4 depends on the geochemical regime and is either biological and/or thermogenic.