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## Experimental simulations of mixed gas hydrates dissociation in response to temperature changes in Qilian Mountain permafrost, China

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Gas hydrates are ice-like crystalline solids consisting of water and gas (predominantly methane). The methane gas bound in hydrate structures and their worldwide occurrence make them interesting not only as a potential energy source but also as a possible climate-relevant factor. Estimations predict that a certain amount of atmospheric CH<sub>4</sub> may originate through dissociation of global gas hydrates, which may exacerbate global warming (Ruppel and Kessler, 2017). In turn, climate warming is not only directly affecting the hydrate distribution, but also perturbing the hydrate stability field, leading to the release of CH<sub>4</sub> from hydrate-bearing sediments. Gas hydrates, particularly those associated within or below shallow permafrost, are likely to be affected by the climate processes. For instance, gas hydrates in Qilian Mountain permafrost (QMP) are found below thin permafrost layers at a shallow depth of around 133–396 m. They might be vulnerable to dissociation due to global warming resulting in a possible higher CH<sub>4</sub> gas emission in this area. Considering the environmental effect, a proper understanding of hydrate dissociation behavior under specific conditions is important for the stability of natural gas hydrate deposits with respect to climate change.

This study focuses on the potential dissociation process of gas hydrates in QMP. Before the observation of hydrate dissociation, mixed gas hydrates are synthesized from pure water and gas mixtures containing CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, CO<sub>2</sub> at conditions close to those in QMP (3.0 MPa, 278 K) with respect to feed gas composition, pressure and temperature. Formed hydrate crystals are analyzed in x-y-z directions applying confocal in situ Raman spectroscopic measurements to identify structures and guest compositions. The dissociation process is based on the thermal conduction simulating global warming and the results are discussed under several isobaric conditions. The Raman spectra continuously record changes in the hydrate phase for each selected crystal over the whole dissociation period. Preliminary results show that the Raman peak intensities for all components start to decrease when the temperature approaches 287 K, indicating the release of gas from hydrate structures. Interestingly, the varying hydrate composition for the measured crystals suggests a heterogeneous dissociation behavior of each single crystal. The results indicate a faster release of CH<sub>4</sub> molecules from the hydrate phase than other components. In addition, the Raman signals of CH<sub>4</sub> gas molecules that trapped in large cages of sII hydrate disappear first

during the dissociation process. After a limited time, mixed gas hydrates decompose completely without evidence of self-preservation effects. These results provide essential information for the estimation of possible methane release from this area in response to future climate warming.

Ruppel, C. D., and J. D. Kessler (2017). The interaction of climate change and methane hydrates, *Reviews of Geophysics*, 55,126-168.