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Clumped isotopes as a tool to discern the sources and sinks of methane in a subglacial environment

Getachew Agmuas Adnew, Thomas Röckmann, Thomas Blunier, Christian Juncher Jørgensen, Malavika Sivan, Sarah Elise Sapper, Maria Elena Popa, Carina van der Veel, and Jesper Riis Christiansen

1Forest, Nature and Biomass, Department of Geoscience and Nature Management, University of Copenhagen, Denmark
2Institute for Marine and Atmospheric research Utrecht, Utrecht University, the Netherlands
3Physics of the Ice, Neils Bohr Institute, University of Copenhagen, Denmark
4Arctic Environment, Department of Ecoscience, Aarhus University, Denmark

Recent observations have revealed that the subglacial meltwater from the western margin of the Greenland Ice Sheet (GrIS) is a net source of methane (1,2). In the future, the Arctic region might become an important methane source due to its vulnerability to anthropogenic warming, glacier retreat, and thawing permafrost.

Mapping the sources of methane and understanding its controlling mechanisms in the subglacial environment of the GrIS are essential for predicting its potential as a climate amplifier and determining its significance in the global carbon cycle. Previous studies used the bulk isotopic composition of methane ($\delta^{13}$C(CH$_4$) and $\delta$D(CH$_4$)) to understand the source and underlying cycling processes of methane below the GrIS (1,2). However, the bulk isotopes have overlapping isotope signatures from microbial, thermogenic, and abiotic sources. In addition, methanotrophic oxidation can modify the isotopic composition, making it challenging to distinguish the source of methane under the GrIS. Using only the bulk isotopes of methane, it is not possible to distinguish aerobic from anaerobic oxidation processes.

Clumped isotopes are measures of how the distribution of heavy isotopes over the various isotopologues deviates from the expected random or stochastic distribution. Measuring the clumped isotopes of methane provides additional constraints to investigate the turnover of methane in the environment. When the methane is at thermodynamic equilibrium, clumped isotopes provide the formation temperature and when the methane is out of thermodynamic equilibrium, the clumped signatures can be used to identify various kinetic gas formation/consumption and fractionation processes that are not possible to reconstruct from the bulk isotopic composition alone.

In this study, we will present the first data on clumped isotope composition of subglacial methane. We will discuss how this new data is used to detail our understanding of the source and sink pathways of subglacial methane. Furthermore, we will show the potential of measuring the clumped isotopes of methane in discerning environmental conditions and types of methanotrophs.
(oxidation pathways).

- Christiansen et al. (2021). DOI: 10.1029/2021JG006308
- Lamarche-Gagnon et al. (2019). DOI: 10.1038/s41586-018-0800-0