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Excess methane, ethane, and propane production in Greenland ice core samples and a first characterization of the $\delta^{13}\text{C}\text{-CH}_4$ and $\delta\text{D}\text{-CH}_4$ signature

Content

Air trapped in polar ice provides unique records of the past atmospheric composition ranging from key greenhouse gases such as methane (CH_4) to short-lived trace gases like ethane (C_2H_6) and propane (C_3H_8). Interpreting these data in terms of atmospheric changes requires that the analyzed species accurately reflect the past atmospheric composition.

Comparisons of Greenland CH_4 records obtained using different extraction techniques revealed discrepancies in the CH_4 concentration for the last glacial. Elevated methane levels (excess methane or $\text{CH}_4(\text{exs})$) were detected in dust-rich ice core sections measured by discrete melt extraction techniques pointing to an artefact sensitive to the measurement technique.

We analyzed Greenland ice core samples for methane and other short-chain alkanes (ethane and propane) covering the time 12-42 kyr using a classic wet extraction technique. The artefact production happens during the melting and extraction step (in extractu) and reaches 14-91 ppb $\text{CH}_4(\text{exs})$ in dusty ice samples. For the first time in ice core analyses, we document a co-production of excess methane, ethane, and propane (excess alkanes) with the observed concentrations for ethane and propane exceeding, at least by a factor of 10, their past atmospheric concentration. Independent of the produced amounts, excess alkanes were produced in a fixed molar ratio of approximately 14:2:1, indicating a common production. We also discovered that the amount of excess alkanes scales generally with the amount of mineral dust (or Ca^{2+}) within the ice samples. Applying the Keeling-plot approach we are able to isotopically characterize $\text{CH}_4(\text{exs})$ revealing a relatively heavy carbon isotopic signature of -46.4‰ ($\pm 2.4\text{‰}$) and a light deuterium isotopic signature of -318.3‰ ($\pm 52.9\text{‰}$) of the excess methane in the samples analyzed.

The co-production ratios of excess alkanes and the isotopic composition of excess methane allows us to confine potential formation processes. We discovered that this specific alkane pattern is not in line with an anaerobic methanogenic origin but indicative for abiotic decomposition of organic matter as also found in sediments.

From the present-day state of research little is known about this process and there is urgent need to improve our understanding for future ice core measurements. Moreover, the already existing discrete records of atmospheric CH_4 in Greenland ice need to be corrected for excess CH_4 contribution ($\text{CH}_4(\text{exs})$, $\delta^{13}\text{C}\text{-CH}_4(\text{exs})$, $\delta\text{D}\text{-CH}_4(\text{exs})$) in dust-rich intervals. While the size of the excess methane production has little effect on reconstructed radiative forcing changes of CH_4 in the past, it is in the same range as the Inter-Polar Difference (IPD) for CH_4 . Knowing the empirical relation of $\text{CH}_4(\text{exs})/\text{Ca}^{2+}$ and $\text{CH}_4(\text{exs})/\text{C}_2\text{H}_6$ allows us to derive a first-order correction of existing CH_4 data sets to revise previous interpretations of the relative contribution of high latitude northern hemispheric CH_4 sources based on the IPD.

Primary author: Ms MÜHL, Michaela (Oeschger Centre for Climate Change Research, and Climate and Environmental Physics, University of Bern)

Co-authors: FISCHER, Hubertus (Climate and Environmental Physics and Oeschger Centre for Climate Change Research, University of Bern); SCHMITT, Jochen (Climate and Environmental Physics and Oeschger Centre for Climate Change Research, University of Bern); BROOK, Edward J. (College of Earth, Ocean, and Atmospheric Sciences, Oregon State University); EDWARDS, Jon S. (College of Earth, Ocean, and Atmospheric Sciences, Oregon State University); LEE, James E. (Los Alamos National Laboratory, Earth Systems Observation); SETH, Barbara (Climate and Environmental Physics and Oeschger Centre for Climate Change Research, University of Bern); BLUNIER, Thomas (Centre for Ice and Climate, Niels Bohr Institute, University of Copenhagen)

Presenter: Ms MÜHL, Michaela (Oeschger Centre for Climate Change Research, and Climate and Environmental Physics, University of Bern)

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