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Coupled artefact production of methane, ethane, and propane in polar ice cores

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Air inclusions trapped in polar ice provide unique records of the past atmospheric composition ranging from key greenhouse gases to short-lived trace gases like ethane and propane. Provided the analyzed species concentrations and their isotopic fingerprints accurately reflect past atmospheric composition, valuable constraints can be put onto biogeochemical cycles. However, it is already known that not all drill sites or specific time intervals are equally suitable to derive artefact-free gas records; e.g., CO₂ data from Greenland ice is overprinted by CO₂ 'in situ' production due to impurities in the ice, and only the cleaner Antarctic ice allows to reconstruct past atmospheric CO₂.

Until recently, CH₄ artefacts in polar ice were only detected on melt affected samples or for short spikes related to exceptional impurity deposition events (Rhodes et al 2013). However, careful comparison of CH₄ records obtained using different extraction methods revealed disagreements among Greenland CH₄ records and initiated targeted experiments.

Here, we report experimental findings of CH₄ artefacts occurring in dust-rich sections of Greenland ice cores. The artefact production happens during the melt extraction step ('in extractu') of the classic wet extraction technique and typically reaches 20 ppb in dusty stadial ice which causes erroneous reconstructions of the interhemispheric CH₄ difference and strongly affects the hydrogen isotopic signature of CH₄ (Lee et al. 2020). The measured CH₄ excess is proportional to the amount of mineral dust in the ice. Knowing the empirical relation between produced CH₄ and the dust concentration of a sample allows a first-order correction of existing CH₄ data sets and to revise previous interpretations.

To shed light on the underlying mechanism, we analyzed samples for other short-chain alkanes ethane (C₂H₆) and propane (C₃H₈). The production of CH₄ was always tightly accompanied with C₂H₆ and C₃H₈ production at amounts exceeding the past atmospheric background levels derived from low-dust samples. Independent of the produced amounts, CH₄, C₂H₆, and C₃H₈ were produced in molar ratios of roughly 16:2:1, respectively. The simultaneous production at these ratios does not point to an anaerobic methanogenic origin which typically exhibits methane-to-

ethane ratios of $\gg 100$. Such alkane patterns are indicative of abiotic degradation of organic matter as found in sediments.

We found this specific alkane pattern not only for dust-rich samples but also for samples that were affected by surface melting from the last interglacial (NEEM ice core) with low dust concentrations. This implies that the necessary precursor is an impurity also present in low-dust ice and the step leading to the production of the alkanes could then be activated when a sufficient boundary condition is met for the production, e.g. by melt/refreeze of surface snow.