



## Inter-polar difference of $\text{CH}_4$ , $\delta\text{D}(\text{CH}_4)$ and $\delta^{13}\text{C}\text{CH}_4$ during the Holocene

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The past variation of the concentration of atmospheric methane ( $\text{CH}_4$ ), the third most important greenhouse gas after water vapour and carbon dioxide ( $\text{CO}_2$ ), is observed to be generally in phase with the northern summer insolation cycle driven by the precession of the rotation axis of the Earth. However, in the mid-Holocene this regularity breaks down, and atmospheric  $\text{CH}_4$  starts to rise again while the northern summer insolation continues to decline. Despite different attempts to explain this feature (e.g. contrasting hypotheses on early human influences and enhanced emissions in the southern tropics), a clear explanation for the evolution of the atmospheric methane concentration has not been found yet.

One possibility to get more information about the highly under-determined system of the methane cycle is to measure the methane concentration in Greenland and Antarctica and to calculate the relative inter-polar difference (rIPD) of methane, allowing us to draw conclusions about the hemispherical imbalance of the source and sink distribution.

More information on the involved production and sink processes of atmospheric  $\text{CH}_4$  can be gained from the isotopic composition ( $\delta\text{D}$  and  $\delta^{13}\text{C}$ ) of  $\text{CH}_4$ . Each source emits methane of a typical isotope signature, and each sink process leads to a certain isotopic fractionation and thus influences the isotopic composition of atmospheric methane.

To exploit for the first time the full parameter set, we also measured the inter-polar difference of  $\delta\text{D}$  ( $\text{IPD}_{\delta\text{D}}$ ) and  $\delta^{13}\text{C}$  ( $\text{IPD}_{\delta^{13}\text{C}}$ ) of methane over the Holocene. The  $\text{IPD}_{\delta\text{D}}$  record results from a high resolution  $\delta\text{D}$  record from the NGRIP (Greenland) ice core and a coarse resolution record of EDML (Antarctica) ice core samples. The NGRIP  $\delta\text{D}$  data show a clear covariation with the long-term  $\text{CH}_4$  concentrations changes during the Holocene. The  $\delta\text{D}$  variations of 8-10 ‰ are significantly larger than our measurement error of 2.3 ‰ which is of significantly better precision than previous data. To avoid any systematic error, the EDML data were measured on the same system and during the same measurement campaign as the NGRIP samples. The resulting  $\text{IPD}_{\delta\text{D}}$  is constant within the measurement error at approximately -16.2 ‰ (north-south) during the entire Holocene. The  $\delta^{13}\text{C}$  measurements (NGRIP and Talos Dome) were done on another system with an measurement error of 0.13 ‰ supporting a long-term decrease in  $\delta^{13}\text{C}$  over the entire Holocene as previously observed (Sowers, QSR, 2010). Our new measurements show that also the  $\text{IPD}_{\delta^{13}\text{C}}$  appears to be relatively constant over the last 12'000 years as well. Given the significant signal in both isotopes of methane and in the methane concentration over the Holocene, the rather constant offset between Greenland and Antarctica is surprising and helps to constrain past changes in the global methane cycle.