



What can we learn about an early human influence on the global methane cycle from bipolar atmospheric CH₄, δD(CH₄) and δ¹³CCH₄ measurements during the Holocene

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The past variation of the concentration of atmospheric methane (CH₄) 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 CH₄ starts to rise while the northern summer insolation continues to decline. Despite different attempts to explain this feature (e.g. contrasting hypotheses on early human influences or enhanced natural emissions in the southern tropics), an unambiguous explanation of the evolution of the atmospheric methane concentration has not yet been found.

In addition to the inter-polar difference (IPD) of methane, allowing us to draw conclusions about the hemispheric imbalance of the source and sink distribution, the isotopic composition (δD and δ¹³C) of atmospheric CH₄ provides further information about the methane cycle. 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 the full parameter set, we measured the methane isotopes on ice cores from both polar regions (NGRIP from Greenland; EDML and Talos Dome from Antarctica) and are able to calculate the inter-polar difference of δD (IPD_{δD}) and δ¹³C (IPD_{δ¹³C}) of methane over the Holocene. To avoid systematic errors, the samples from both hemispheres have been measured on the same system and during the same measurement campaign for each parameter. The NGRIP δD data show a clear covariation with the long-term changes in CH₄ concentrations during the Holocene. The δD variations of 8-10 ‰ are significantly larger than our measurement error of 2.3 ‰. However, the resulting IPD_{δD} is constant within the measurement error at approximately -16.5 ‰ (north-south) during the entire Holocene. The δ¹³C records (with a measurement precision of 0.13 ‰) show a clear decrease in δ¹³C of about 1.9 ‰ and an IPD_{δ¹³C} of about -0.3 to -0.5 ‰ over the early part of the Holocene and stable δ¹³C values for the last 5'000 years with an IPD_{δ¹³C} of -0.7 ‰.

To obtain a better understanding of the processes involved in the methane cycle we assume the sink strengths (or atmospheric lifetime) and the hemispheric mixing terms to be constant, allowing us to do a unique box model inversion, which provides integrated CH₄ emissions with the weight-averaged isotope signatures for both hemispheres. The inversion shows the larger changes in the methane emission rate and the corresponding isotopic signature in mid-Holocene to occur in the southern hemisphere. This may be in disagreement with the hypothesis of early human influences, since rice agriculture with their CH₄ emissions are assumed to be allocated mainly in the northern tropics at that time. Further, we use atmospheric box models to assess different CH₄ emission scenarios to constrain the past changes in the global methane cycle.