

## Atmospheric CH<sub>4</sub> concentration during the Holocene reconstructed from the NEEM (Greenland) and Dome Fuji (East Antarctica) ice cores

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Methane (CH<sub>4</sub>) is an important greenhouse gas, whose atmospheric concentration has been increasing due to human activities for the last few centuries. Orbital-scale variations of atmospheric CH<sub>4</sub> correlate with climatic precession, because the size of wetlands and their CH<sub>4</sub> production rate respond to Northern Hemisphere (NH) summer insolation, through the variations in temperature and rainfall on NH landmasses. The correlation between CH<sub>4</sub> concentration and NH summer insolation held for the last three interglacial periods, but the relationship collapsed during the latter half of the Holocene. NH summer insolation kept decreasing whereas the atmospheric CH<sub>4</sub> concentration increased since ~5 kyr BP. Several explanations have been proposed for this trend, such as peat growth in circum-Arctic region<sup>1</sup>, emission from tropical wetlands due to increasing rainfall in the Southern Hemisphere (SH)<sup>2,3</sup> and agricultural activity<sup>4</sup>, and the exact mechanisms have been under continuing debate.

Inter polar difference (IPD) of CH<sub>4</sub> concentrations have provided important constraint on the evolution of CH<sub>4</sub> source distribution and its relationship with climate<sup>2,5,6,7</sup>. However, time resolution and analytical precision of previous studies have not always been adequate to investigate precise IPD. In addition, reconstruction of accurate CH<sub>4</sub> variation is difficult during most of the Holocene from Greenland ice cores, because the depths for this time period often corresponds to poor quality ice (brittle zone). To reconstruct the CH<sub>4</sub> IPD during the Holocene, we have been measuring CH<sub>4</sub> concentrations in the NEEM (Greenland) and Dome Fuji (DF) (Antarctica) ice cores. Accurate CH<sub>4</sub> reconstruction from the Holocene NEEM ice core is challenging because of the brittle zone. We indeed found high CH<sub>4</sub> spikes in the brittle zone, thus we investigated them by measuring additional 3–5 samples from the neighboring depths (within ~50 cm, ~5 years) and checking the reproducibility, and then rejected the data which is more than 15 ppb higher than their means. Reproducibility after removing the outliers are ±2.5 and ±1.7 ppb for the NEEM and DF ice cores, respectively.

We investigate the integrity of our Holocene CH<sub>4</sub> data. The CH<sub>4</sub> variations of the NEEM core, including centennial to millennial variations in the brittle zone, agree well with the GISP2 data (recent high-precision data by the Oregon State University group) (ref.7 and unpublished data). For Antarctica, the variations of CH<sub>4</sub> concentration of the DF core also agree well with those of the WAIS divide core from West Antarctica (ref.7 & 8 and unpublished data), after considering centennial-scale smoothing effect on the DF record caused by slow gas trapping. These comparisons suggest that our new records, as well as the most recent records by other groups, provide reliable reconstruction of the past atmospheric CH<sub>4</sub> variations over the entire Holocene.

IPD are deduced from two different combinations of cores: the NEEM and DF cores, and the NEEM and WAIS Divide cores. For this analyses, the gas time scales of the NEEM and DF cores are placed on the WAIS Divide ice core chronology by pattern matching of the CH<sub>4</sub> records. IPD from both NEEM/DF combination and NEEM/WAIS combination increased from the early Holocene to mid Holocene, and then

decreased toward the late Holocene. We employ a simple 3-box model<sup>2,6</sup> to deduce CH<sub>4</sub> emissions from different latitudinal bands at 1000-yr intervals. The model calculates the emissions in the low-latitude box (30°S-30°N) and northern box (30-90°N), while small emission from southern box (90-30°S) is kept constant. The model results show that northern emission decreased, while low-latitude emission increased during the last half of the Holocene. This suggests significant contribution from the low-latitude sources to the atmospheric CH<sub>4</sub> increase since ~5 ka. A recent model study suggests that CH<sub>4</sub> emission from the SH tropics may have increased due to SH summer insolation rise<sup>7</sup>. Several terrestrial proxies suggest increased rainfall in the tropical regions in South America during the latter half of the Holocene<sup>9</sup>, probably in response to the increase in SH summer insolation. Although we cannot reject the anthropogenic hypothesis at this stage and more investigations are needed, our results are consistent with the hypothesis that tropical SH emission was responsible for the CH<sub>4</sub> rise during the latter half of the Holocene.

1)Blunier et al., 1995. 2)Chappellaz et al., 1997. 3)Singarayer et al., 2011. 4)Ruddiman & Thomson., 2001. 5)Nakazawa et al., 1993. 6)Brook et al., 1996. 7)Mitchell et al., 2013. 8)WAIS Divide Project Members, 2014. 9)Prado et al., 2013.

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