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Changes in the atmospheric CH_4 gradient between Greenland and Antarctica during the Last Glacial and the transition to the Holocene

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Abstract. Significant variations in the interpolar difference of atmospheric CH_4 concentration over the Holocene period were observed by *Chappellaz et al.*, [1997]. Here we extend this study to the Last Glacial and the transition to the Holocene. We observe a gradient of -3±4 parts per billion by volume (ppbv) during the Last Glacial Maximum. It increases to 26±10 ppbv during the Bølling/Allerød and remains at 26±9 ppbv during the Younger Dryas cold period. On average, we find an interpolar difference of 14±4 ppbv during the cold phases and of 37±10 ppbv during the warm periods of the Last Glacial. With a three-box model we derive from the measured gradients the contributions of methane from the Tropics and the mid-to-high latitudes of the northern hemisphere. The Tropics have been the largest source in all glacial epochs. The contribution by the northern latitudes have been very small during the last glacial maximum but surprisingly large during the earlier part of the glacial epoch. The model result suggests completely unexpected, that the higher atmospheric CH_4 concentration during the warm Dansgaard/Oeschger events are caused by a higher source strength of the northern latitudes and not of the Tropics.

1. Introduction

Methane is a radiatively and chemically active trace gas with a variety of natural and anthropogenic sources [Chappellaz et al., 1993b; Hein et al., 1997]. Changes in the concentration of CH_4 has two potential causes: a variation in the intensity of the global CH_4 source, and/or a change in the magnitude of the major CH_4 sink, the reaction with OH radicals. Model calculations show that, globally, the OH concentrations were 10 to 30% higher than the present values for both the preindustrial Holocene and the Last Glacial (LG) [Thompson, 1992 and references therein]. Therefore, changes in the CH_4 source strength have to be mainly responsible for variations of the atmospheric CH_4 concentration for both epochs. Before the industrial revolution, the dominant methane sources were natural wetlands [Chappellaz et al., 1993a]. Based on the Greenland CH_4 record, the fast fluctuations parallel to temperature changes in Greenland, so called Dansgaard-Oeschger (D-O) events during the LG were explained tentatively with variations of the extent and activity of tropical wetlands. Combining CH_4 records from Greenland and Antarctica over the same time scale allows us to extend the determination of the interhemispheric gradient into the last glacial period. This pole-to-pole gradient of a few percents results from the uneven distribution of sources and sinks over the latitudes. Sources are mainly terrestrial and thus, exist primarily in the northern hemisphere, while the sinks are distributed roughly uniformly over the globe. This additional information allows us, together with a box model, to quantify roughly the latitudinal distribution of CH_4 sources.

2. Methane time series

The investigations are based on CH_4 records from the European "GReenland Ice-core Project" (GRIP) ice core drilled in Central Greenland (Summit: 72°34'N, 37°37'W), from one drilled at Byrd Station, West Antarctica (Byrd: 79°59'S, 120°01'W) and from one drilled at Vostok, East Antarctica (78°28'S, 106°48' E).

Essential for the determination of interhemispheric differences are reliable time scales for the involved ice cores. The methane records show fast and drastic variations parallel to (D-O) events during the second half of the last glacial period. These variations were used to synchronize the Byrd and the Vostok ice cores to the GRIP time scale [*Blunier et al.*, 1998]. The relative uncertainty of the CH_4 records reduces to 200 years, much lower than the relative uncertainty applying the original individual time scales and good enough for our purpose. We base our analysis on the synchronized time scales by *Blunier et al.*, [1998].

If $\delta^{18}O$ values of the ice (a proxy for air temperature) are compared with methane measurements it is important to know the age difference between the gas and the ice. The air, which gets enclosed and isolated in bubbles at 50-150 m below snow surface, has a mean age which is younger than the surrounding ice. The age difference depends mainly on temperature and accumulation rate. This difference can be calculated accurately for the GRIP core with an uncertainty of about ±300 years during the LG [*Schwander et al.*, 1997].

The measurements for the methane records shown in Figure 1 have been done in Bern and Grenoble. The two laboratories have calibrated their measurements on the same standard and also compared measurements on ice samples from the same depth [*Sowers et al.*, 1997]. For a description of the extraction and measurement procedure see *Chappellaz et al.*, [1997].

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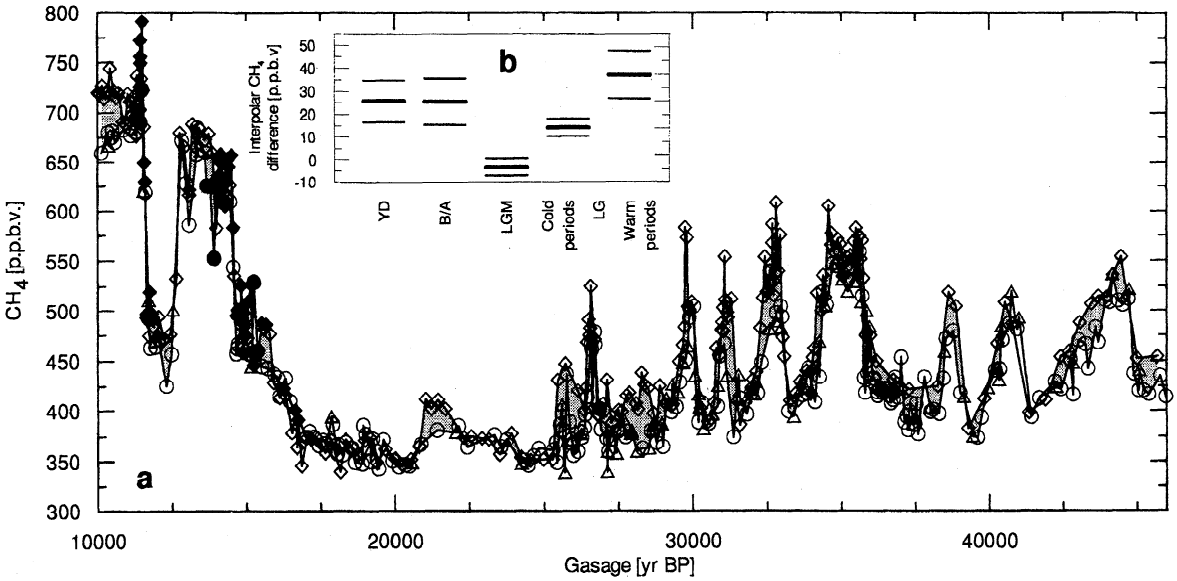

Figure 1. (a) Comparison of the Greenland and Antarctic mean CH_4 concentrations from 46-9.5 kyr. Antarctica: solid line with circles and triangles; Byrd station: open circles, published data [*Blunier et al.*, 1998]; filled circles, new data. Vostok: open triangles, published data [*Blunier et al.*, 1998]. GRIP: solid line with diamonds; open diamonds, published data [*Blunier et al.*, 1998]; filled diamonds, new data. (All new data were analyzed in Bern.)
(b) Interpolar CH_4 difference and 1σ error bars for five selected time intervals, warm and cold stages during the Last Glacial (LG), the Last Glacial Maximum (LGM), the Bølling/Allerød (B/A) and the Younger Dryas (YD) period.

3. Greenland/Antarctic CH_4 Difference

Figure 1a shows new data along with previously published data. The mean temporal resolution could be improved to 132 years for the GRIP record and to 170 years for the Byrd record due to the new measurements. For the Vostok record only published results are used with a mean temporal resolution of 250 years. The results of the Byrd and Vostok records have been combined to one Antarctic record. The temporal resolution and the accuracy of the synchronization is not sufficient to reconstruct a continuous record of the interhemispheric atmospheric methane concentration difference. Therefore, distinct climatic epochs have been selected and the mean interhemispheric difference for the corresponding epoch calculated by subtracting the mean Antarctic concentration from the mean Greenland concentration. Mean concentrations and standard errors for each interval and for each record have been calculated weighing the data points by the time interval they represent.
The mean values and the differences for the four epochs: Younger Dryas (YD), Bølling/Allerød (B/A), Last Glacial Maximum (LGM) and the glacial epoch with Dansgaard-Oeschger (D-O) events are shown in Table 1. One mean value for the whole time interval 25,000-46,000 years BP with warm and cold phases is not satisfactory. The sampling resolution on the other hand does not allow us to determine the interhemispheric gradient for each individual D-O event and the intermediate cold stages. A main difficulty is to determine the start and termination of warm and intermediate cold events. One possibility is to use the $\delta^{18}O$ record of the GRIP ice core. However, for this approach we have to take into account the age difference between the enclosed gases and the surrounding ice. We replaced the single value of the $\delta^{18}O$ record with a spline with a cut-off frequency of 620 years (four times the mean resolution of all methane data from 46-25 kyr BP). All intervals above the average of -40.25 ‰ were selected to belong to warm, all intervals below this value to cold phases (Figure 2). The differences between Greenland mean values and Antarctic mean values fluctuate between 18 and 70 ppbv for warm periods and between -28 and 35 ppbv for cold periods. The differences for the single periods have

Table 1. Characteristics of Greenland and Antarctic CH_4 records over selected time intervals.

Time Interval	YD	B/A	LGM	Cold periods	Warm periods
(kyr BP)	11.63-12.5	12.7-14.5	16.7-20.3	25.0-46.0	25.0-46.0
Greenland Number of data points	11	23	19	57	80
Mean CH_4 concentration (ppbv)	485	656	362	419	502
Standard deviation of the mean (ppbv)	3.7	5.9	2.6	3.2	5.0
Antarctica Number of data points	8	17	24	117	82
Mean CH_4 concentration (ppbv)	459	630	365	405	465
Standard deviation of the mean (ppbv)	8.3	8.2	2.7	2.2	9.1
Interpolar difference and 1 σ uncertainty (ppbv)	26±9	26±10	-3±4	14±4	37±10

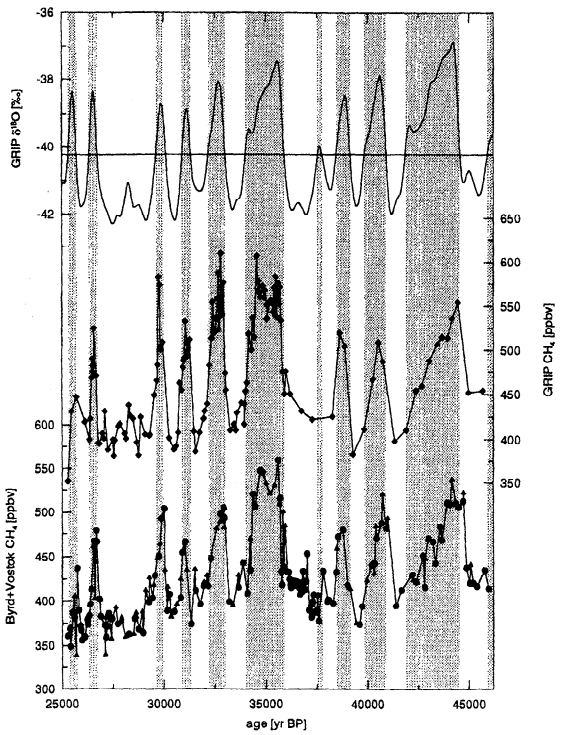

Figure 2. Time intervals for the warm periods and the cold period. Top, splined $\delta^{18}O$ record with mean level. (The spline is the mean of all splines we got from a Monte Carlo simulation. For the Monte Carlo simulation, 1000 runs were simulated. Each run takes into account the fact that the deviations from the mean values show a Gaussian distribution and calculates a smoothed spline with a cutoff period of 620 years [*Enting*, 1987]). Shaded area, time intervals for the warm periods. Middle, GRIP CH_4 record (mean concentration black diamonds). Bottom, Antarctic CH_4 record (Byrd, mean concentration black dots; Vostok, mean concentration black triangles).

large uncertainties and cannot be interpreted in terms of environmental changes. Therefore, we calculated a more significant mean difference for all warm periods together and for all cold periods together. The mean difference for all warm periods is 37±10 ppbv, for all cold periods 14±4 ppbv.

The uncertainty of the limits of all other time intervals due to the air-ice age difference is negligible. We have made a Monte Carlo simulation taking into account the uncertainty of the time scales regarding the grouping criterion. CH_4 values for warm and cold periods were calculated varying the age for a CH_4 data point randomly within the uncertainty limits and applying the grouping criterion afterwards.

Compared to the initial value, the mean values of these runs decreases for the warm periods and increases for the cold periods in both hemispheres. This is not surprising as the synchronization of the time scales was done by finding a maximum correlation between $\delta^{18}O$ and CH_4. Despite the uncertainty in the absolute values for GRIP and Byrd CH_4 records, the interpolar difference changed within the estimated error (14±4 ppbv during the cold periods and 37±6 ppbv during the warm periods).

The results concerning the interhemispheric differences between the northern and the southern hemisphere for different climatic periods are summarized: The differences between 46-25 kyr BP have been 14±4 ppbv during cold periods and 37±10 ppbv during warm periods. The difference decreased at the beginning of the LGM to -3±4 ppbv and increased to 26±10 ppbv in the B/A. The difference remained practically unchanged (26±9 ppbv) during the YD and reached a level of 44±4 ppbv [*Chappellaz et al.*, 1997] at the beginning of the pre-boreal.

Brook et al., [1999] have analyzed the CH_4 gradient for the YD (18±5 ppbv), B/A (22±7 ppbv), LGM (18±7 ppbv) and a single warm event (D-O event 8) (40±10 ppbv)based on the GISP2 and the Taylor Dome (Antarctica) records. The deviation is during all events within the error limits except for the LGM. However, *Brook et al.*, [1999] analyzed the CH_4 gradient for the time interval from 18-26 kyr BP while we cover the time interval from 16.7-20.3 kyr BP. This together with the lower resolution of their record is likely to explain the discrepancy between the two results.

4. Source Distribution

Using a three-box model [Chappellaz et al., 1997] and the CH_4 concentrations for the selected time intervals quantitative contributions of CH_4 sources in the tropics (30°S-30°N) and the middle to high latitudes of the northern hemisphere (30°N-90°N) were calculated for steady state conditions. The box model was developed and applied for the Holocene by Chappellaz et al., [1997]. They used the CH_4 distribution for the years 1984-1990 [Steele et al., 1992] and an estimate for the source [Thompson et al., 1993] and sink distribution [Fung et al., 1991]. With these modern conditions the transport time between the boxes and the lifetime in each box was obtained. Both 2D and 3D model calculations show that, globally, the present and the last glacial OH concentrations are 10 to 30% lower than the values in the period from 1200 to 1800 AD [Thompson, 1992]. The global sink term is therefore increased by 20%. Finally, since only values for the two polar boxes are available from ice cores, the CH_4 concentration in the tropic box was determined to get a source of 12 Tg/yr during the LGM and the cold periods respectively of 15 Tg/yr during the other time intervals in the southern box, consistent with source estimates for this region [Fung et al., 1991]. The main assumptions of the model are an unchanged interhemispheric transport time, an unchanged sink distribution and a sink term homogeneously increased by 20% compared with present-day conditions over the whole period [Chappellaz et al., 1997].

5. Results and Discussion

The calculated source distribution among the three boxes is presented in Figure 3 for five time intervals. While it was earlier assumed that the boreal sources were relatively small and rather constant during the last glacial epoch our results suggest substantial contributions from northern latitudes with variations between the various climatic periods on the same order as for the tropics.

Only during the LGM a low contribution from the northern latitudes (14 Tg/yr), comparable to the southern latitudes contribution (12 Tg/yr), is observed. The contribution from the northern latitudes more than triples during the transition from the LGM to the mild B/A and remains, unexpected, at an

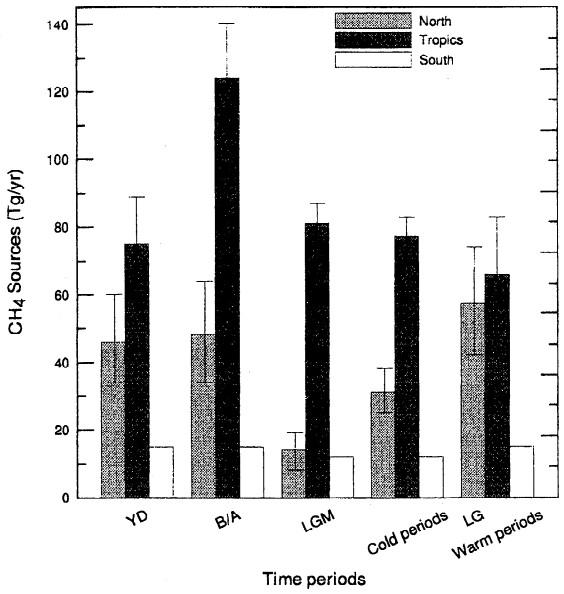

Figure 3. Source distribution among the three boxes of our model for each of the selected time intervals (warm and cold stages during the Last Glacial (LG), the Last Glacial Maximum (LGM), the Bølling/Allerød (B/A) and the Younger Dryas (YD) period). The source in box South was fixed at 12 Tg/yr during the LGM and the cold periods respectively at 15 Tg/yr during the other time intervals.

about constant level during the cold YD period. This despite the fact that the YD had its largest impact in the northern hemisphere. The increase of the atmospheric CH_4 concentration between LGM and B/A can be attributed to source increases in the tropics and in the northern latitudes at about equal parts. The decrease between B/A and YD is primarily attributed to a source decrease in the tropics.

Even more surprising are the variations of the source distribution during the glacial epoch before the LGM. The increases of the atmospheric CH_4 concentration during the D-O events is due to changes in the source strength in northern latitudes. The source strength in the tropics is even slightly smaller during D-O events as during the cold periods. However, this result is due to the large errors still very uncertain. This uncertainty will have to be reduced by further measurements. We are also aware that it is not satisfactory to use mean values for various D-O events together. The source distribution is not necessarily the same for individual events. The spectacular results obtained by the mean values will be motivation enough to measure high resolution records for single events in the future.

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References

Blunier, T., J. Chappellaz, J. Schwander, A. Dällenbach, B. Stauffer, T. Stocker, D. Raynaud, J. Jouzel, H.B. Clausen, C.U. Hammer, and S.J. Johnsen, Asynchrony of Antarctic and Greenland climate change during the last glacial period, Nature, 394, 739-743, 1998.

Brook, E.J., J. Severinghaus, S. Harder, and M. Bender, Atmospheric methane and millennial scale climate change, AGU Monograph on Mechanisms of Millennial Scale Climate Change, 112, 165-175, 1999.

Chappellaz, J., T. Blunier, S. Kints, A. Dällenbach, J.-M. Barnola, J. Schwander, D. Raynaud, and B. Stauffer, Changes in the atmospheric CH_4 gradient between Greenland and Antarctica during the Holocene, J. Geophys. Res., 102, 15987-15999, 1997.

Chappellaz, J., T. Blunier, D. Raynaud, J.M. Barnola, J. Schwander, and B. Stauffer, Synchronous changes in atmospheric CH_4 and Greenland climate between 40 and 8 kyr BP, Nature, 366, 443-445, 1993a.

Chappellaz, J.A., I.Y. Fung, and A.M. Thompson, The atmospheric CH_4 increase since the Last Glacial Maximum, Tellus, 45 B, 228-241, 1993b.

Enting, I.G., On the Use of Smoothing Splines to Filter CO_2 Data, J. Geophys. Res., 92, 10977-10984, 1987.

Fung, I., J. John, J. Lerner, E. Matthews, M. Prather, L.P. Steele, and P.J. Fraser, Three-dimensional model synthesis of the global methane cycle, J. Geophys. Res., 96, 13033-13065, 1991.

Hein, R., P.J. Crutzen, and M. Heimann, An inverse modeling approach to investigate the global atmospheric methane cycle, Global Biogeochem. Cycles, 11, 43-76, 1997.

Schwander, J., T. Sowers, J.-M. Barnola, T. Blunier, B. Malaizé, and A. Fuchs, Age scale of the air in the summit ice: Implication for glacial-interglacial temperature change, J. Geophys. Res., 102, 19483-19494, 1997.

Sowers, T., E. Brook, T. Blunier, A. Fuchs, M. Leuenberger, J. Chappellaz, J.M. Barnola, and D. Etheridge, An inter-laboratory comparison of techniques for extracting and analyzing trapped gases in ice cores, J. Geophys. Res., special issue, 1997.

Steele, L.P., E.J. Dlugokencky, P.M. Lang, P.P. Tans, R.C. Martin, and K.A. Masarie, Slowing down of the global accumulation of atmospheric methane during the 1980s, Nature, 358, 313-316, 1992.

Thompson, A.M., The Oxidizing Capacity of the Earth's Atmosphere: Probable Past and Future Changes, Science, 256, 1157-1165, 1992.

Thompson, A.M., J.A. Chappellaz, I.Y. Fung, and T.L. Kucsera, The atmospheric CH4 increase since the Last Glacial Maximum, 2, Interactions with oxidants, Tellus, 45 B, 242-257, 1993.

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