Atmospheric Methane in Ice Cores

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Abstract The reconstruction of air trapped in ice cores provides us the most direct information about atmospheric CH₄ variations in the past history. Ice core records from the "Three Poles (Antarctica, Greenland and Tibetan Plateau)" reveal the detailed fluctuations of atmospheric CH₄ concentration with time and are allowed to quantify the CH₄ differences among latitudes. These data are indispensably in the farther study of the relationship between greenhouse gases and climatic change, and of the past changes in terrestrial CH₄ emissions. Ice cores reconstruction indicates that atmospheric CH₄ concentration has increased quickly since industrialization, and the present day's level of atmospheric CH₄ (1800 pp-bv) is unprecedented during the past Glacial-Interglacial climate cycles.

Key words Ice core, Methane record, Climate change.

1 Introduction

Anthropogenic emission has resulted in momentous change in atmospheric CH₄ concentration since the industrialization. It is urgent for us to understand how the global climate and ecosystem will response to this change, vice versa, how the changing global climate and ecosystem will influence the geochemistry cycles of the CH₄. Ice core records involve the complicated interactions among all the systems (terrestrial ecosystem, ocean, atmosphere chemistry, climate, etc.), at the same time, reveal their feedbacks to climate variation with time. The ingredients and their concentrations in the atmosphere have been documented by direct measurements only since several decades ago, whereas the extraction and analysis of air occluded as bubbles in ice provides the most promising method for accurately reconstructing the concentration history of atmospheric trace gases, and this method can trace the evolution of atmospheric trace gases back to, at least, 740 ka ago (EPICA Community Members 2004).

The air occluded as bubbles in ice core has been recognized since the first attempt

of deep ice core drilling at Byrd Station and Ross Ice-shelf in the Geophysical Year 1957-1958 (Patenaude 1959; Ragle 1960). So far, large numbers of ice cores have been recovered in the Antarctica and Greenland. These cores have been or are being planed to be used for reconstructing the atmospheric greenhouse gases. Ice cores drilled from very high elevations on the Tibetan Plateau, such as the Dasuopu ice core (7200 m a. s. l) (Thompson et al. 2000; Xu et al. 1999) and Muztagh Ata ice core (7010 m a. s. l) provide a unique opportunity for obtaining sub-tropical latitude CH₄ reconstruction. The past 2 ka variation of CH₄ concentration of the sub-tropical latitude has been documented for the first time (Xu et al. 2001; Yao et al. 2002).

2 Atmospheric CH₄ variation in the glacial-interglacial cycles

The past climatic change has been described as the repeating glacial-interglacial of around 100 ka cycles since the Late Quaternary (the last 1,000 ka) (Imbrie 1992), and is just like the record revealed by the Vostok ice core from the Antarctica covering the last 420 ka (Petit et al. 1999). This ice core reconstruction indicates that the main trends of CH₄ concentration are similar over the last four climatic cycles, i. e. with the maximum during interglacial epochs (650-770 ppbv) and the minimum during glacial periods (320-350 ppbv) (Figure 1). The overall remarkable correlation of CH₄ with Antarctic temperature suggests that the greenhouse gases are important as amplifiers of the initial orbital forcing (Raynaud et al. 2000), and also reflects the interaction between terrestrial ecosystem and climatic change. The processes occurring at the scale of the glacial-interglacial changes are generally not directly relevant to the recent and future century scale variability of the climate. But the overall correlation between greenhouse trace-gases and climate over glacial-interglacial cycles highlights the potential of past records to investigate the sensitivity of climate to greenhouse gases under different climatic conditions and provides tests for climate models intended to simulate future responses to increasing concentrations of greenhouse gases.

The global warming contribution due to the initial forcing caused by direct effect of increasing greenhouse gases during the glacial-interglacial transitions is assessed to be about 0.95°C (Petit et al. 1999). This initial forcing should be amplified by the succedent rapid feedbacks, such as the correlative water vapor and albedo modifications. The effect of increasing greenhouse gases due to anthropogenic emission does also as the case. Studies (Berger et al. 1998; Weaver et al. 1998) assumed that the greenhouse gases and their correlative rapid feedbacks have a significant contribution (possibly 2-3°C) to the global averaged glacial-interglacial temperature increase.

Atmospheric CH₄ concentration had been changed significantly which was correlated tightly to climatic cycles during the past 160 ka (Chappellaz et al. 1990; Chappellaz et al. 1993a). Chappellaz et al. (1990; 1997) investigated the past natural variations of atmospheric CH₄ in principle, and suggested that the sub-tropical terrestrial sources intensity dominated atmospheric CH₄ variations, and global CH₄ level reflects the changes in the extent of wetlands linked to the changes of the precipitation and temperature in low latitude regions.

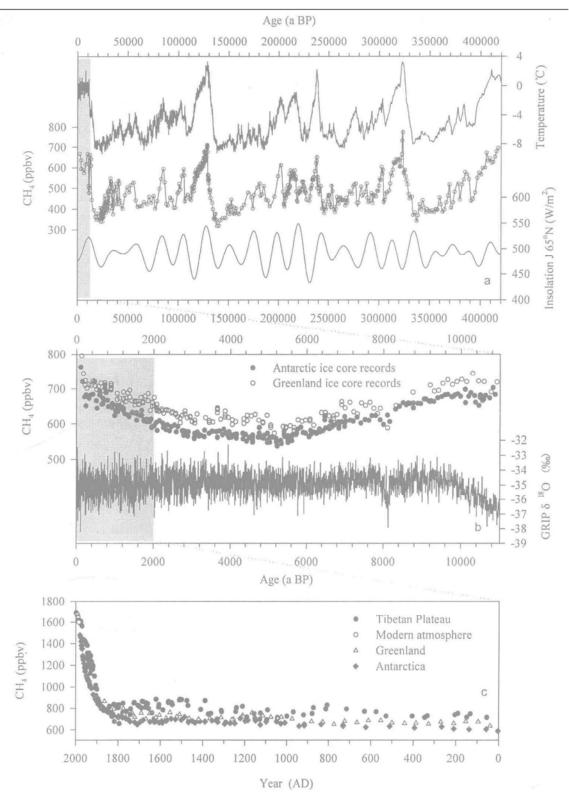


Fig. 1 Ice core CH₄ records in different time scales. a; Vostok ice core record of CH₄ and temperature over the past 420 ka (Petit et al. 1999); b; Atmospheric CH₄ gradient between North and South recorded in ice cores during the Holocene (Chappellaz et al. 1993b, 1997; Raynaud et al. 1988; Blunier et al. 1993, 1995); c; Atmospheric CH₄ record over the past 2 ka (Xu et al. 2001; Yao et al. 2002; Blunier et al. 1993; Dlugokencky et al. 1994; Etheridge et al. 1992, 1998).

The ice core records of the past atmospheric CH₄ concentration reveal several most striking features. Changes of the past atmospheric CH₄ concentration from Antarctic ice cores showed large variations in glacial-interglacial climate cycles. The CH₄ concentrations were only about 350 ppbv during the glacial periods and around 700 ppbv during the interglacial epochs. At one time, there existed decipherable fluctuations of CH₄ concentration which coincide with 20 ka periodicity of the climatic changes, such as the obvious increase of atmospheric CH₄ in the 80 ka BP and 100 ka BP warm periods. During the glacial, interglacial events were familiar, and accompanied by relative large increase of atmospheric CH₄ concentration. In Greenland ice core records these interglacial events showed a feature as quickly warming and slowly cooling, and then the temperature abruptly decreasing to glacial level (Chappellaz et al. 1993b). There were about 150 ppbv atmospheric CH₄ increase in these interglacial events at least during the past 42 ka.

There is a remarkable positive correlation between the atmospheric CH₄ concentration and the temperature reflected by stable isotope in the Vostok record (Figure 1a), and the overall coefficient (r²) is 0.73, this suggests that atmospheric CH₄ can play an important role as an amplifier in the initial orbital forcing during the glacial-interglacial climatic cycles, and then leads to a sequence of climate forcing as the decreasing albedo resulted from retreating ice cover and other possible changes (Raynaud et al. 1993; Lorius et al. 1990).

3 Atmospheric CH₄ during the Holocene epoch

The study of Holocene is specific for understanding the interactions between climate change and geochemical cycles, and thereby provides us the possible feedback processes related to the future global warming (Raynaud et al. 2000). The both high resolution CH4 records in Greenland and Antarctic ice cores covering the whole Holocene epoch are now available (Chappellaz et al. 1993b, 1997; Raynaud et al. 1988; Blunier et al. 1993, 1995). The ice core records show that the natural variability of atmospheric CH₄ concentration is within 150 ppbv prior to anthropogenic emission, and the Greenland ice core record is averagely 6% higher than the Antarctic reference. However, the present day's interpolar CH4 gradient is almost four times of the Holocene's, predominatly due to anthropogenic sources in the northern hemisphere (Dlugokencky et al. 1994). The similar air bubble formation processes, characterized by similar resolution, were found both in Greenland ice cores (GRIP and GISP2) and Antarctica ice cores (D47, EPICA, and Byrd), and thereby they can be used for direct comparison study (Figure 1b). The lowest inter-polar CH₄ gradient of 33±7 ppby is found during 5-7 ka BP interval, whereas the highest CH₄ difference of 50 ± 3 ppbv occurred in 2.5-5 ka BP, the relative large interpolar gradient is also observed during the Early Holocene. CH4 variations in the Early Holocene display the epilogue scene of the warming process which the climate evolved from cold in the Last Glacial to warm of interglacial. Atmospheric CH₄ concentration increased abruptly, and the inter-polar CH4 gradient enlarged synchronously at the end of the Younger Dryas (YD), during which the warming process was interrupted near its end. This phenomenon indicates that the mid-latitudes or the boreal regions in the north hemisphere as potential CH₄ sources, with the climate warming, begin to play an important role. A detailed investigation of CH₄ variations during the Last Interglacial suggested that the abrupt CH₄ degassing from hydrate reservoirs (Brook *et al.* 2000) and the boreal wetlands expanding (Raynaud *et al.* 2000) were probably the main contributor of atmospheric CH₄. The determination of the latitudinal gradients of CH₄ trapped in ice cores under different climate conditions can provide us a tool for better understanding of the latitudinal distribution of the CH₄ wetland sources.

Although the Holocene climate is relative stable, the startling fluctuation of 150 ppbv of atmospheric CH₄ concentration is observed all the same. The whole Holocene can be divided into three periods based on the CH₄ variation trends: 10 ka BP-5 ka BP, 5 ka BP-3 ka BP and 3 ka BP- present. The early Holocene was characterized by high atmospheric CH₄ concentration, which decreased to the lowest level at around 5 ka BP. Atmospheric CH₄ kept stable from 5 ka BP to 3 ka BP, and then increased steady-going to the pre-industrialized level (Figure 1b). It is perplexing that the decrease of atmospheric CH₄ concentration in the middle Holocene and the subsequent slowly increase, excluding the sharp CH₄ decrease event at 8. 2 ka BP, were not correlative with the temperature changes recorded in ice core (Figure 1b). This indicates that the correlations between the climate changes and the greenhouse gases are complicated. The CH₄ fluctuations recorded in ice cores testify that the terrestrial ecosystem and the hydrological cycle have experienced remarkable changes during the Holocene.

4 Atmospheric CH₄ in the past 2 ka

The high resolution atmospheric CH₄ concentrations coving the past 2 ka from the Dasuopu ice core in the Tibetan Plateau and from the Polar Regions are now available as shown in the Figure 1c. The overall trend of sub-tropical CH₄ recorded in the Dasuopu core shows a slight increase during the Late Pre-industrial Holocene. The most important feature of the Dasuopu ice core record is that the CH₄ concentration and temporal variation amplitude are larger than those in the polar ice core records, and the average CH₄ concentration in the Dasuopu core is 782 ppbv, the maximum temporal variation exceed 200 ppbv which has never been found in polar ice records for the 1850 years (0-1850 AD). The high resolution and intense natural fluctuations provide a unique opportunity for investigating the CH4 latitudinal variations and terrestrial emissions. There exist three periods of 0-600 AD, 900-1400 AD and 1750-1850 AD, which characterized obviously by lower CH₄ concentrations as shown in Figure 1c. The corresponding CH₄ concentrations for the three periods are 735 ppbv, 746 ppbv and 776 ppbv, which are 47 ppbv, 36 ppbv and 6 ppbv lower respectively than the average concentration of 782 ppbv during the Late Pre-industrial Holocene of the past 2 ka. Although the averaged CH₄ concentration during 1750-1850AD is only 6 ppbv less than its Late Pre-industrial Holocene's averaged level, the highest concentration of 880 ppbv in the Dasuopu ice core has never been found in the Polar Regions and the lowest concentration of 658 ppbv which is equivalent to the coexistent Antarctic record are both occurred in this period that makes the natural fluctuation of atmospheric CH₄ concentrations the largest, i. e. 220 ppbv or 28% of the average CH₄ concentration of the Late Pre-industrial Holocene of the past 2 ka. It is striking that the natural fluctuation is so high, and may be suggested that the sub-tropical terrestrial CH₄ sources had changed singularly with climate variations during such a short period of only 100 years. In the Dasuopu record, the higher averaged CH₄ concentrations of 791 ppbv and 833 ppbv which are 9 ppbv and 51 ppbv higher than its Late Pre-industrial Holocene's level are took place respectively in the time intervals of 600-900 AD and of 1450-1750 AD.

The Dasuopu record indicates that the averaged CH₄ concentration during the Little Ice Age is 806.5 ppbv which is higher than its Late Pre-industrial Holocene's mean level, and even is higher than the relative higher concentration of 791 ppbv occurred in the period of 600-900 AD. But there are obviously three valleys of CH₄ concentration corresponding to the periods of 1400-1450 AD, 1600-1670 AD and 1750-1850 AD when the climates were cold as recorded by ice cores from the Tibetan Plateau (Yao et al. 1997; 2002).

Recently, increased attention has been paid to the impacts of anthropogenic emissions on greenhouse gases due to their contribution to global warming. Although the earlier study (Rasmusen and Khalil 1984) of ice core records from Antarctica indicated that the obvious impact of anthropogenic emission on the atmospheric greenhouse gases started around 1850 AD, the most subsequent ice cores suggested a remarkable impact started from 1750 AD (Craig and Chou 1982; Etheridge et al. 1992, 1998; Stauffer et al. 1985; Steele et al. 1992). Our Dasuopu record shows an unprecedented and continuous CH₄ increase started around 1850 AD with a concentration increase of 1000 ppbv in about 120 years.

The Tibetan Plateau, Greenland and Antarctica represent the three typical latitudes of sub-tropical region and high latitudinal region in the north hemisphere and high latitude of the south hemisphere respectively. Due to the oceans, we assume that the intensity of CH₄ sources in the south hemisphere have been stable at least since the Holocene. The primary CH₄ contributions came from the low-latitudinal wetlands and the boreal peatlands (Chappellaz et al. 1997; Fung et al. 1991; Dallenbach et al. 2000), so the ice core CH₄ records from the Tibetan Plateau and Greenland can reflect sensitively the changes of their latitudinal sources, and also the atmospheric CH₄ transfers.

The Antarctic Law Dome cores (Etheridge et al. 1998) combined with D47 (Blunier et al. 1993) core and Greenland GRIP core (Blunier et al. 1993) combined with Eurocore (Etheridge et al. 1998) provide very detailed CH₄ reconstruction for the last 2 ka. Figure 1c shows the CH₄ concentrations from the Dasuopu core, Greenland cores, and Antarctic cores. The average difference is 66 ppbv with Greenland and 107 ppbv with Antarctica, and the average difference between Greenland and Antarctic records is 44 ppbv in the time interval of 0-1850 AD. The larger CH₄ gradients between those in the Dasuopu and the Polar Regions suggested that the low-middle latitude acted as the most important CH₄ source in the Late Pre-industrial Holocene. A detailed comparison can be made in the following four periods: 0-900 AD,

900-1450 AD, 1450-1750 AD and 1750-1900 AD.

Larger CH₄ gradients of 127 ppbv and 84 ppbv between sub-tropical latitudes and the southernmost and northernmost region respectively have been found in the first 900 years, even though the earliest century is the coldest period recorded by the δ¹⁸O in the Dasuopu ice core (Yao et al. 2002). Corresponding to the warmest period, the maximum difference of 165 ppbv /and 150 ppbv between the Dasuopu record and Antarctic /and Greenland record is occurred around 800 AD, at the same time, the smallest gradient of about 15 ppbv is appeared between Greenland and Antarctica. It is also characterized by larger CH4 differences between low-middle latitudes and the Polar Regions in the forepart (1450-1750 AD) of the Little Ice Age. The Dasuopu δ^{18} O profile indicates that 700-900 AD is the warmest period in the past 2 ka (Yao et al. 2002). Though there is no signal in the Dasuopu record can prove that the forepart (1450-1750 AD) of the Little Ice Age is also a warmer period, but the glacier accumulation (precipitation) is very high in this period recorded by the Quelccaya ice core (Thompson et al. 1985) and the Dasuopu ice core (Duan et al. 2002). We, therefore conclude that Monsoon evolution incorporated with high CH_4 input in the south Asia might be responsible for the relatively high CH₄ concentration observed in the Dasuopu ice core in the above-mentioned two periods.

Smaller CH₄ gradients between low-middle latitude and the Polar Regions appeared in 900-1450AD, especially in the 19th century when climate was cold. In the period of 900-1450AD, the average CH₄ difference is 76 ppbv between the Dasuopu and Antarctica, and 34 ppbv between the Dasuopu and Greenland, but there is a relatively larger interpolar CH₄ difference (average of 42 ppbv), especially in the Medieval Warm Period, the interpolar difference enlarged to about 55 ppbv. The smallest difference of CH4 mixing ratio between low-middle latitude and the Polar Regions has been found in the 19th century, the average difference is 28 ppbv with Antarctica, and -39 ppbv with Greenland, but the inter-polar difference reached its maximum of 67 ppbv. It is evident that boreal sources became increasingly important, potentially due to the already significant contribution of human-induced sources. Based on the Modern Monitoring Network, Dlugokencky et al (1994) found that the CH₄ concentration in the northern hemisphere is 143 ppbv higher than the southern hemisphere in recent times, due presumably to large increases in anthropogenic emissions from northern hemispheric sources. Rasmussen and Khail (1984) studied the inter-hemispheric gradient of recent atmospheric CH₄ concentration. They also found an evident gradient between the Arctic and the Antarctica, and the CH₄ concentration over the Arctic is 10% higher than that over Antarctica.

To demonstrate the pole to pole distribution of CH₄ on a long time scale, Blunier et al (1995) have implemented an ice core CH₄ reconstruction for the Holocene and revealed that there were significant changes in the interpolar difference of CH₄ mixing ratio with time. It is 44 ppbv for the whole Holocene. According to Dlugokencky's study (1994) based on modern monitoring network, the CH₄ concentration in atmosphere decreases from the northernmost to the southernmost. However, the CH₄ concentration reconstructed for the past 2 ka from the Dasuopu ice core is highest among all the ice core reconstruction. One possible interpretation of the result is that the

strong monsoon convection would bring CH₄ from the south Asia, particularly India, up to the Dasuopu Glacier. One important clue to support the speculation is that the CH₄ concentration in the Dasuopu Glacier was getting higher as climate became warmer, which might imply that the stronger the monsoon, the more CH₄ was brought up to the Dasuopu Glacier by the monsoon system. Chappellaz et al (1997) proposed that global wetlands, particularly the tropical wetland is the major factor controlling global CH4 budget based on the study of CH4 concentration reconstruction of the polar ice cores during the Holocene. The change of the wetland in the tropical regions is mainly controlled by hydrological process, or in other words, by monsoon intensity. High CH₄ concentration will be a consequence of strong monsoon, and vice versa (Street-perrott, 1993). It seems that the Dasuopu ice core is now recording not only the strong anthropogenic input, but also strong natural input by intensified hydrological process, or more specifically by stronger monsoon. The study of Steel et al. (1992) also indicates that the changes in CH₄ emission between 30-90°N are particularly important in the global CH₄ increasing trend. In addition, according to Steel et al. (1992), the average CH₄ concentration (1702. 19 ppbv) in the Northern Hemisphere is higher than global average (1656. 56 ppbv) and Southern Hemisphere (1610. 91 ppbv). On the other hand, the CH₄ concentration in the Northern Hemisphere itself fluctuates from site to site. Locating at 28°N, the Dasuopu Glacier might be a sensitive recorder of low-mid latitude CH4 concentration. To further confirm the feature, more ice core CH₄ reconstructions are necessary in this region.

5 Global CH₄ emission since the Last Glacial Maximum

Atmospheric CH₄ fluctuations reflect as proposed (Chappellaz et al. 1997) the intensity variations of CH₄ sources, and then the CH₄ gradients among different latitudes can indicate latitudinal sources strength. The excellent correlation between atmospheric CH₄ concentrations and temperatures recorded in ice cores from the Polar Regions indicates that the climate change is the dominating factors in controlling the strength variations of CH₄ sources (Raynaud et al. 1993, 2000; Petit et al. 1999; Chappellaz et al. 1990). A comparison study on the south Asian monsoon precipitations and atmospheric CH₄ fluctuations over the past 160 ka recorded in the Vostok ice core had been executed by Chappellaz and others (Chappellaz et al. 1990), the authors suggested that the monsoons play an important role in controlling the atmospheric CH₄ concentration variations by influencing the extent of the low-latitudinal wetlands. It is obvious that the emission strength of low-latitudinal CH₄ sources predominates over the global CH₄ concentration. The CH₄ concentration in the north hemisphere is higher than in the south hemisphere, and at the same time the low-mid latitude CH₄ concentration in the north hemisphere is the highest over the globe prior to industrialization, that is indicated by the CH₄ gradients of 107 ppbv, 66 ppbv and 44 ppbv respectively between the Dasuopu record and Antarctica record, the Dasuopu record and Greenland record, as well as Greenland record and Antarctic record. Since the industrialization, anthropogenic CH_4 emission has been the primary sources, and

nowadays the pole to pole gradient expanded to 143 ppbv which has never been found in the past (Dlugokencky et al. 1994).

The latitudinal distribution of CH₄ sources (concentrated in northern boreal and tropical areas) had largely changed since the Last Glacial Maximum (LGM) that was confirmed based on a simple three-box model by the combined analysis of ice cores from Greenland and Antarctica (Chappellaz et al. 1997; Dallenbach et al. 2000) (Figure 2). Still, this analysis of available CH4 data from Greenland and Antarctica relied on several critical assumptions, one of them is that the CH₄ sources between 30 and 90°S remain unchanged with time. The model output shows that the major contribution of atmospheric CH4 lies on the tropical latitudes, and it is surprising that the boreal region appears to be also important especially during the Early Holocene and the Late Holocene (Chappellaz et al. 1997). The total terrestrial CH₄ sources of 30°S-90°N are 196 and 154 Tg/a respectively, and in which 58 ± 10 and 71 ± 7 Tg/a fractions are contributed by those of 30°N-90°N during the time intervals of 0.25-1 ka BP and 2.5-5 ka BP, that means the contributions from the tropical areas (30°S-30°N) reach respectively 128 and 83 Tg/a. The above results suggest that with the growing contribution of tropical sources the boreal sources such as peatlands have became decreasingly important since the mid-Holocene. The boreal source (30°N-90°N) was very weak during the LGM, and it was only 14 Tg/a which is equivalent to the contribution from the latitudes of 30°S - 90°S. However, the boreal sources increased by 3 times and reached 48 Tg/a during the warming period of the last glacial-interglacial transition (Bolling/Allerok), and the tropical sources were also quite increased from 81 Tg/a to 124 Tg/a in this period. The abrupt cold event of YD has an important impact on the sources distribution of the north hemisphere, the boreal source still kept its B/A higher level, whereas the tropical source decreased to 75 Tg/a which is even lower than its Last Glacial level. In generally, the tropical area had been the primary source of atmospheric CH₄ from the Last Glacial to the Holocene; especially during the LGM the global sources were concentrated in these areas. The YD cold event made an exception, the boreal source remained higher emission (Dallenbach et al. 2000), on the contrary the tropical source decreased to its glacial level that resulted in the global average CH₄ concentration also decreased to glacial level.

Up to now, we are lack of the knowledge about the exact distribution of CH₄ sources spatio-temporal variation, the oxidized depletion of atmospheric CH₄ in the past histories, and the possible sources, among them the most possibility is the misjudgment of CH₄ sources in the south hemisphere. So it is an urgent task for building of the past CH₄ data sets from low-mid latitudes.

6 Summary

The ice core records not only the climate changes, but also the greenhouse gases fluctuations. In order to fully understand the phase relation on various time scales between climate change and greenhouse gases, it is a vital project to study the ice-air age difference in different climatic conditions, at the same time, the technology of stable isotopes analysis on air trapped in ice should be well developed for the under-

standing of the gases sources.

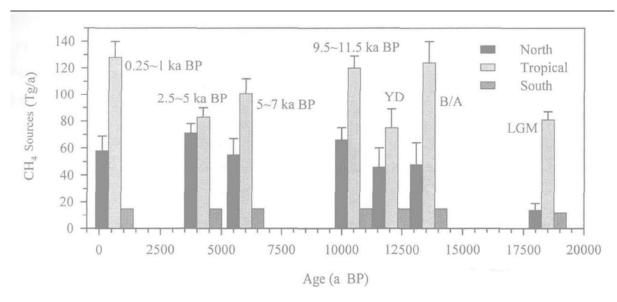


Fig. 2 Latitudinal distribution of methane sources since the Last Glacial Maximum (Chappellaz et al. 1997; Dallenbach et al. 2000).

Atmospheric CH₄ variations keep in phase with climate changes on the orbital time scale, but this relation was broken during the Holocene. Atmospheric CH₄ fluctuations are relative to terrestrial ecosystem and aero-chemical processes, CH₄ concentration is mainly dominated by the sources strength. Different climate patterns cause different feedbacks of the oceans, terrestrial ecosystems and aero-chemical processes, and then induce the variations of atmospheric CH₄ concentration. The complicated relations between climate and CH₄ indicate that the mechanisms are different in the strength changes of CH₄ sources driven by climate changes on different scales, and vice versa.

The comparison study of the past 2 ka CH₄ records in ice cores from the "Three Poles" allows quantify for the first time the difference of CH₄ mixing ratio between polar and sub-tropical latitudes during pre-industrial time, and we are cognizant of the complexity of the correlations between greenhouse gases and climate change from this practice. Several critical assumptions on the global distributions of CH₄ sources which are constructed based on the polar data sets are now faced with challenge.

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