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Trend and seasonal variations of atmospheric CH_4 in Beijing

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Abstract: The atmospheric CH_4 in Beijing is still increasing, even though its increasing rate has significantly decreased from 1.76%/a during 1985—1989 to 0.50%/a during 1990—1997. The seasonal variation of CH_4 concentration showed a double-peak pattern, one peak appearing in winter and the other in summer. It is evident that the annually seasonal variations of atmospheric CH_4 in Beijing are different. From 1986 to 1997, the atmospheric CH_4 increased by 185 ppbv, 37% and 21% of which were due to the increase in winter and in summer, respectively. After 1993, the annually seasonal increasing rate of CH_4 concentration in summer (due to emission from biogenic sources) is negative while the increasing rate in winter (due to emission from non-biogenic sources) is positive about 25 ppbv/a. As a result, the increase of CH_4 emission from non-biogenic sources in winter is the major reason that caused the annually seasonal increasing rate from 1993 to 1997. The biogenic sources in Beijing are shrinking while the non-biogenic ones (such as fossil fuel combustion) are enlarging.

Key words: atmospheric CH_4; increasing rate; seasonal variation; annually seasonal variation

Introduction

Methane (CH_4) is an important trace organic gas with the highest concentration in the atmosphere. Because CH_4 has strong ability to absorb infrared-red light and then warm the atmosphere, its greenhouse effects are closely following those of carbondioxide (CO_2). The main biogenic sources of atmospheric CH_4 include rice fields, natural wetlands, ruminants and termites. The main non-biogenic source (Wang, 1993) include biomass combustion and fossil fuel exploitation and utilization. The active organic carbon, microbic anaerobiosis and temperature mainly control biogenic sources. Non-biogenic sources are mainly controlled by the exploitation rate of fossil fuels, industrial development and urbanization. Reactions with OH in the atmosphere are the most significant sink of the atmospheric CH_4 (Paul, 1995). CH_4 can remove CFCs from the atmosphere by reacting with Cl. In the past decades, the increasing rate of CH_4 in the atmosphere has been decreased from 17—21 ppbv/a during 1978—1982 to 8—13 ppbv/a during 1988—1990 (IPCC; Houghton, 1994). The increase of CH_4 was correlated to the changes of wetland area and the rates of fossil fuel exploitation and utilization (Houghton, 1994; Khalil, 1989), however, the reason of CH_4 enhancement in the atmosphere remains uncertain.

By cooperating between the Institute of Atmospheric Physics, Chinese Academy of Science (IAP-CAS) and Oregon Graduate Center, USA, the seasonal variation and trend of atmospheric CH_4 in national terrestrial baseline areas (Wang, 1990) were firstly investigated at Minqin of Gansu Province, China during 1985—1987. The project, monitoring the atmospheric CH_4 and its long-term variation in Beijing, was carried out by IAP-CAS. The observations were expected to help understand the relationship between urban ecosystem with intensive disturbance and long-term global change of atmospheric CH_4, and to reduce the uncertainties on the reason of the increase of the atmospheric CH_4.

1 Experimental method

Sampling instruments were installed on the third floor of the meteorological observation tower in IAP-CAS. No higher building and local source existed within the range of 1 km^2. Duplicate samples were taken between 14:00 to 15:00 PM when the air was mixed so well that the samples represented the situations of free troposphere. Air samples were taken once or twice a week. Each sample was split equally into three for duplicate analysis. The CH_4 concentration in the sample was calculated by those average-analyzing values. Air was sampled with specially designed stainless steel flasks. Their inner surfaces were electropolished to avoid gas adsorption. Moreover, with no leakage valves, composition changes of samples stored in those kinds of flasks are not monitored

within several months. The samples were analyzed with HP5890 II Gas Chromatography (GC) with Flame Ionization Detector (FID) in IAP-CAS. NIST of USA and NSC of China provided the standards samples. The precision of CH_4 concentration analysis was 0.05%—0.10%. The detection limit, stability of instrument system and the analysis precision, met the criterions for measuring atmospheric CH_4. Therefore, the data presented in this paper are comparable.

2 Results and discussion

The data measured in the past decades showed that the atmospheric CH_4 in Beijing is still increasing with significant seasonal, monthly and annually seasonal variation.

2.1 Annual variation

The long-term observation on atmospheric CH_4 in Beijing started in 1985. The average annual concentration and the increasing rates are listed in Table 1. The temporal variation is shown in Fig.1. It is obvious that: (1) The atmospheric CH_4 in Beijing has been increasing, but the increasing rate decreased significantly (Fig.2). The highest increasing rate was observed between 1985—1989, which is 1.76%/a, almost 23 ppbv/a increased. But the rate is down to 0.5%/a, only 9.46 ppbv/a increased 1990—1997. (2) From 1985 to 1997, the atmospheric CH_4 in Beijing was 1865 ± 68 ppbv. But it increased to 1915 ± 73 ppbv during the past five years. In 1997, it reached 1944 ± 75 ppbv, which is much higher than the national terrestrial baseline and the global average level.

Table 1 Average concentration and inter-annually increasing rate of the atmospheric CH_4 in Beijing

Year	Average concentration, ppbv	STDS, ppbv	Sample numbers, groups	Increase value, ppbv	Increase rate, %*	Average increase rate, %**
1985	1.722×10^3	26	45			
1986	1.760×10^3	58	96	37	2.14	
1987	1.795×10^3	55	71	35	2.00	1.76
1988	1.835×10^3	52	102	40	2.19	
1989	1.862×10^3	63	93	27	1.48	
1990	1.876×10^3	63	84	14	0.73	
1991	1.885×10^3	111	32	9	0.49	
1992	1.889×10^3	83	90	4	0.21	
1993	1.908×10^3	79	34	19	1.01	
1994	1.928×10^3	74	49	20	1.05	0.50
1995	1.917×10^3	56	67	−11	−0.59	
1996	1.928×10^3	80	89	11	0.59	
1997	1.944×10^3	76	77	16	0.81	
Average	1.865×10^3	68	71	18	1.10	1.01

*. percentage of difference of two continuous years to their average; **. average increasing rate of simulated values

The average concentrations of atmospheric CH_4 from some national terrestrial baseline areas (Minqin) are listed in Table 2. The average value 1918 ± 57 ppbv in Beijing is about 82 ppbv higher than that in Minqin. And the standard deviation of observed data in Beijing is about 17 ppbv higher than that in Minqin. However, before 1992, about 40 ppbv is higher in Beijing than that in Minqin, with its standard deviation 42 ppbv higher. Meanwhile, either the annually average or the monthly average standard deviation is significantly lower than the present data in Beijing. It implies that natural ecosystems in Beijing have been suffering more intensive anthro-disturbance. The difference between urban and terrestrial baseline areas is getting greater. This appearance could also be explained by the results: the data observed from Dinghushan and Xinglong Field Station, which are both near developed areas, are much higher than that observed from

Table 2 Average CH_4 concentration in baseline areas of China

Station	Concentration, ppbv	STDS, ppbv
Xinglong	1.845×10^3	30
Waliguan	1.802×10^3	8
Dinghushan	1.907×10^3	54
Changbaishan	1.789×10^3	67
Average	1.836×10^3	40

The data are from reference Wang, 1996

Waliguan Station, which is far away from developed areas. With similar geological and climatic conditions to Minqin Station, the data from Waliguan Station are a little higher than that from Changbaishan Station. But the standard deviation of Waliguan is the lowest, 8.30 ppbv, which is very close to the value observed at Minqin, 12 ppbv. Because the concentration in Beijing is 4.5% higher than that in terrestrial baseline areas, Beijing should be a stronger source. Furthermore, both of them are respectively 8% and 3% higher than the global average concentration 1776 ppbv (estimated from reference; Houghton, 1994). It implies that China might be a potential source of global atmospheric CH_4.

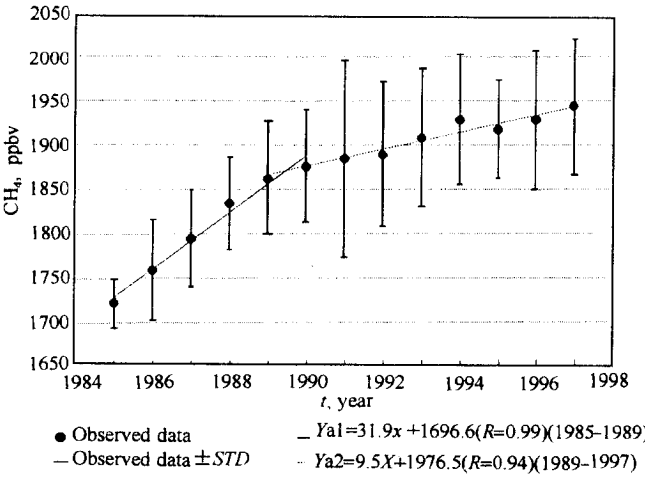

Fig. 1 Concentration and variation of atmospheric CH_4 in Beijing

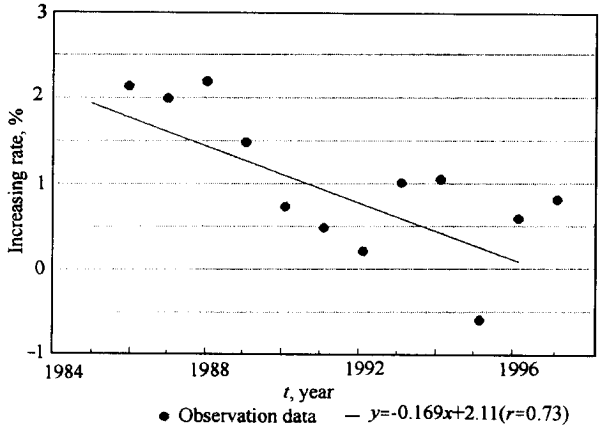

Fig. 2 Variation of increasing rate of atmospheric CH_4 in Beijing

2.2 Seasonal variation

As shown in Fig. 3, the increase of atmospheric CH_4 concentration in Beijing has significant seasonal variation. Because of the alternative effects of biogenic and non-biogenic sources, the seasonal variation of CH_4 concentration in the atmosphere of Beijing shows a double peak pattern. One peak appears in winter (December to February), the other in summer (June to September). The winter peak may be caused by the combustion of fossil fuel, especially coal. The summer peak may be caused by biogenic sources of CH_4 emission. Paddy rice fields, swamps and fermentation of organic wastes are important biogenic sources of CH_4. With extension of urban areas and increasing population, millions of tons of life organic wastes are produced in Beijing every year. The decomposition to produce CH_4 of organic wastes is affected by water temperature and anaerobic bacteria. In Beijing, heavy rainfall in summer usually produces a suitable status of anaerobic condition for CH_4 production. Moreover, temperature is so high that many organic substances degrade to CH_4, which is released into air and increases the concentration of CH_4 in the atmosphere.

As shown in Fig. 4, two-peak seasonal pattern appeared annually during the observation period excluding 1986 to 1987, when no obvious peak was observed in winter. From 1986 to 1997, the seasonal average value in summer was about 12 ppbv higher than that in winter. One peak observed in winter and the other in summer. It implies that the impact of biogenic sources on CH_4 in the atmosphere in Beijing was more intensive than that of non-biogenic ones in the past decades. However, the highest peak has been shifted from summer to winter in the past decades as shown in Fig. 4 by the twelve seasonal variation curves. It will be discussed in section 2.4.

2.3 The variation of monthly average concentration

The variation of monthly average concentration of atmospheric CH_4 in Beijing showed a similar

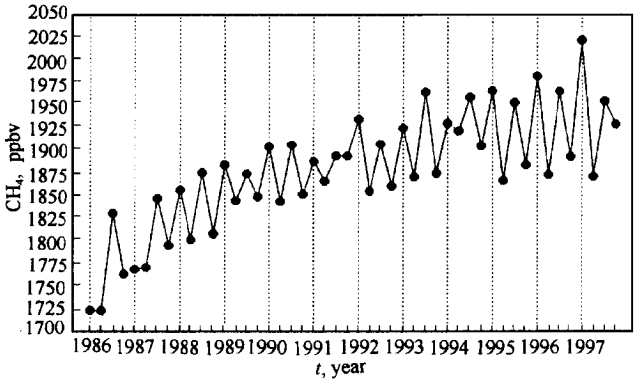

Fig.3 Seasonal variation of atmospheric CH_4 in Beijing (1986—1997)

pattern (Fig.5). Fig.6 shows the average results of Fig. 5. It is clear that the atmospheric CH_4 remains a high level in winter without great fluctuation. It means that the average value of CH_4 concentration, which is mainly from the combustion for warming in winter. In March, the atmospheric CH_4 started to decrease because the combustion for warming stopped. In April, either emission intensity or atmospheric CH_4 dropped to the lowest level. In May, when the temperature increases, CH_4 emissions from biogenic sources were promoted. The atmospheric CH_4 starts to increase and reaches the highest value in July. In August, temperature falls down and the intensity of biogenic sources decreases. As a result, the CH_4 gradually declines. Decrease of atmospheric CH_4 from the highest level in July to the lowest level in October occurres more slowly than the increase from the lowest level in May to the highest one in July (Fig.6). After warming systems starts to run in mid-November, the atmospheric CH_4 increases until the first peak value appears at the beginning of the next year. Alternative influences of biogenic and non-biogenic sources on atmospheric CH_4 account for the double-peak pattern of monthly variation. Although the local meteorological factors may affect the concentration values to the same extent, they do not change the long-term trend and seasonal variation pattern of atmospheric CH_4 in Beijing.

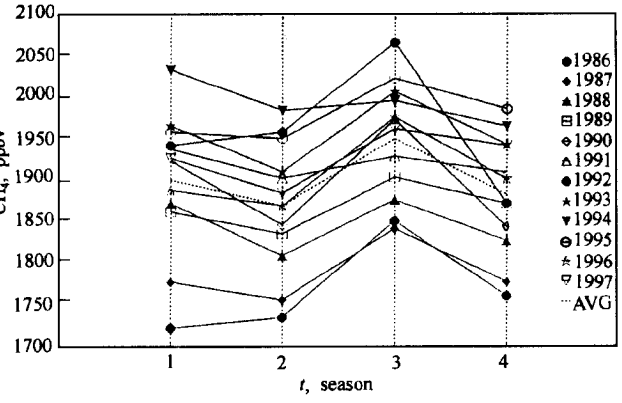

Fig.4 Comparison in seasonal variation of atmospheric CH_4 in Beijing

2.4 Annually seasonal variation

Based on the long-term observation in urban areas with intensive anthropogenic disturbances, it is possible to discuss the impacts of biogenic and non-biogenic sources on the changes of atmospheric CH_4.

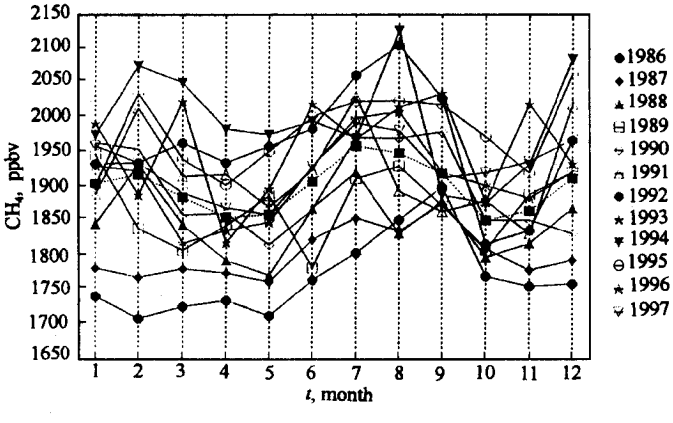

Fig.5 Comparison in monthly variation of atmospheric CH_4 in Beijing

In Fig. 7, the annually seasonal variation of average CH_4 concentration for each season was analyzed with regression method, respectively. The results indicate that the annually seasonal variations of atmospheric CH_4 are different among the four seasons. The variation in winter is the largest, with the highest annually seasonal increasing rate, while those in other seasons are relatively constant, with

constant increasing rate. In Fig. 7(E), the average concentration in winter increases at a rate of 22 ppbv/a or 1.3%/a which is much higher than that of the three seasons, 12 ppb/a or 0.70%/a. From 1986 to 1997, the atmospheric CH_4 increased by 185 ppbv, 37% and 21% of which were due to the increase in winter and in summer, respectively. The increase of atmospheric CH_4 at the highest rate is mainly due to the emissions from non-biogenic sources-combustion of coal and gas for warming in winter.

In the latest five years, the rate of atmospheric CH_4 in summer appears to be negative, as shown in Fig. 8(A). In winter, however, annually seasonal increasing rate is 25 ppbv/a or 1.3%/a (Fig. 8(B)). The highest peak of atmospheric CH_4 has been shifted from summer to winter since 1995. The level of the atmospheric CH_4 in Beijing is mainly controlled by non-biogenic sources instead of biogenic ones. It implies that the decrease of increasing rate of global atmospheric CH_4 is mainly due to two reasons. Firstly, area of biogenic sources could not be extended. Secondly, recent trend of atmospheric CH_4 may be mainly determined by emission intensity of non-biogenic sources. Although main biogenic sources, such as paddy rice fields, livestock, landfill etc., remain their present emission level, the atmospheric CH_4 should be continuously increasing, for further developing of industry and commerce, extending of urban areas and increasing of population.

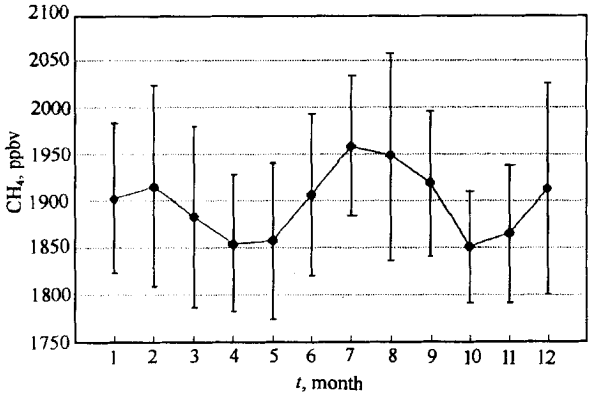

Fig. 6 Variation of average monthly atmospheric CH_4 in Beijing (1986—1997)

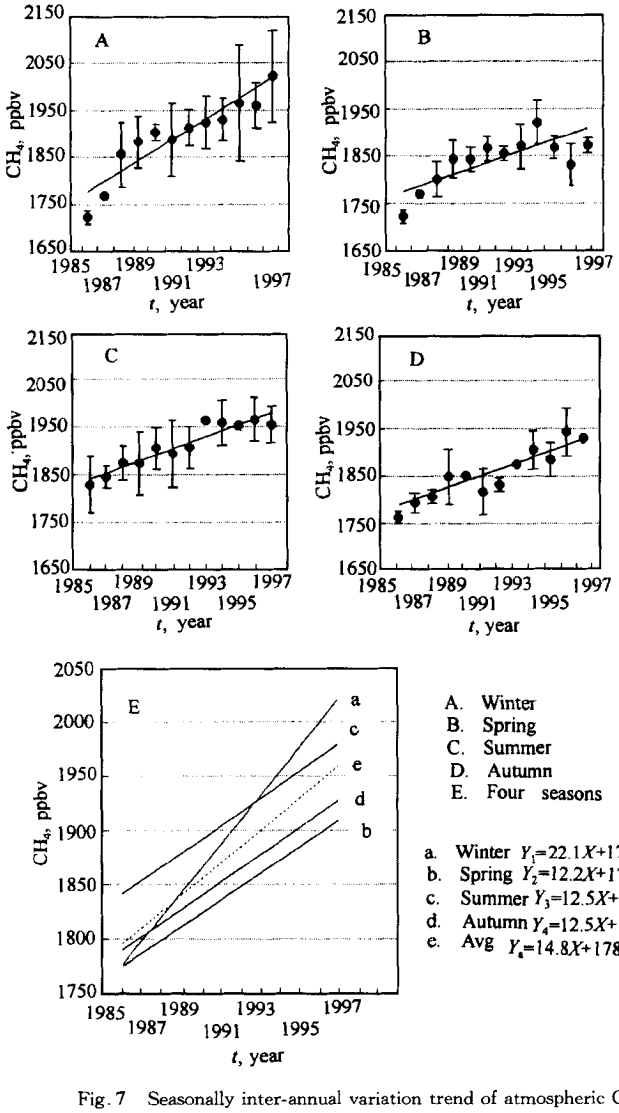

Fig. 7 Seasonally inter-annual variation trend of atmospheric CH_4 in Beijing (1986—1997)

3 Conclusion

Some conclusions could be drawn from above results and discussion. They are:

Because the atmospheric