Proc. NIPR Symp. Polar Meteorol. Glaciol., **8**, 14–18, 1994

RELATIONSHIP BETWEEN ATMOSPHERIC CO_2 AND CH_4 CONCENTRATIONS AT SYOWA STATION, ANTARCTICA

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Abstract: Precise measurements of the atmospheric CO_2 and CH_4 concentrations have been continued at Syowa Station since 1984 and 1987, respectively. Measured concentrations show secular increase, together with seasonal cycle and irregular variations. Negative correlation is clearly seen between the secular trends of the CO_2 and CH_4 concentrations. The increase rates of CO_2 and CH_4 show oscillations with periods of 2.3 to 2.8 years. The phases of the average seasonal cycles of CO_2 and CH_4 coincide with each other, but interannual variations of the observed seasonal cycle of CO_2 are larger than those of CH_4.

1. Introduction

Carbon dioxide (CO_2) and methane (CH_4) are well known as major greenhouse gases, and their atmospheric concentrations have increased for the last 250 years due to increase of human activities. Combustion of fossil fuel is thought to be a main cause of the CO_2 increase. However, the rate of CO_2 increase is highly variable with time and is not necessarily proportional to the amount of CO_2 released from anthropogenic sources (KEELING, 1983). These fluctuations of the rate of CO_2 increase could be attributed to change in CO_2 exchange between the atmosphere and the biosphere and between the atmosphere and the oceans. For better understanding of the global CO_2 cycle, it is necessary to reveal the causes of the fluctuations.

It is thought that the increase of the atmospheric CH_4 concentration is due to rice cultivation, ruminants, landfills, venting of natural gas at wells, pipeline leakage of natural gas, coal mining and biomass burning (CICERONE and OREMLAND, 1988). If atmospheric OH radicals have been decreased secularly, perhaps due to increase of the atmospheric CO concentration by fossil fuel combustion, this effect may also contribute to the CH_4 increase (KHALIL and RASMUSSEN, 1985). However, respective contributions to the concentration increase of CH_4 are not yet understood quantitatively. Therefore, some uncertainties still exist in our understanding of the global budget of atmospheric CH_4. Precise and long

term measurements of atmospheric CH_4 may provide us with important information for solving such a problem.

We have continued systematic measurements of atmospheric CO_2 and CH_4 concentrations at Syowa Station, Antarctica since 1984 and 1987, respectively (TANAKA *et al.*, 1987; NAKAZAWA *et al.*, 1991a; AOKI *et al.*, 1992). In this paper, the results of these measurements are given and compared with each other, especially in terms of the secular trend and the seasonal cycle.

2. Results and Discussion

2.1. CO_2 concentration

In situ measurements of the atmospheric CO_2 concentration have been continued at Syowa Station since February 1984. The location of the station is remote from industrial regions and vegetated continents, and contamination due to station activities is hardly observable. The standard deviation of daily mean CO_2 concentration ranges between 0.01 and 0.15 ppmv. Therefore, a simple statistical data selection scheme (*cf.* NAKAZAWA *et al.*, 1991a) was applied to original data to obtain the background values and only 4.4% of all available data were eliminated as outliers. Figure 1 shows daily mean CO_2 concentrations calculated from the selected data. It is clear from this figure that variations of the CO_2 concentration can be divided into three components: secular trend, seasonal

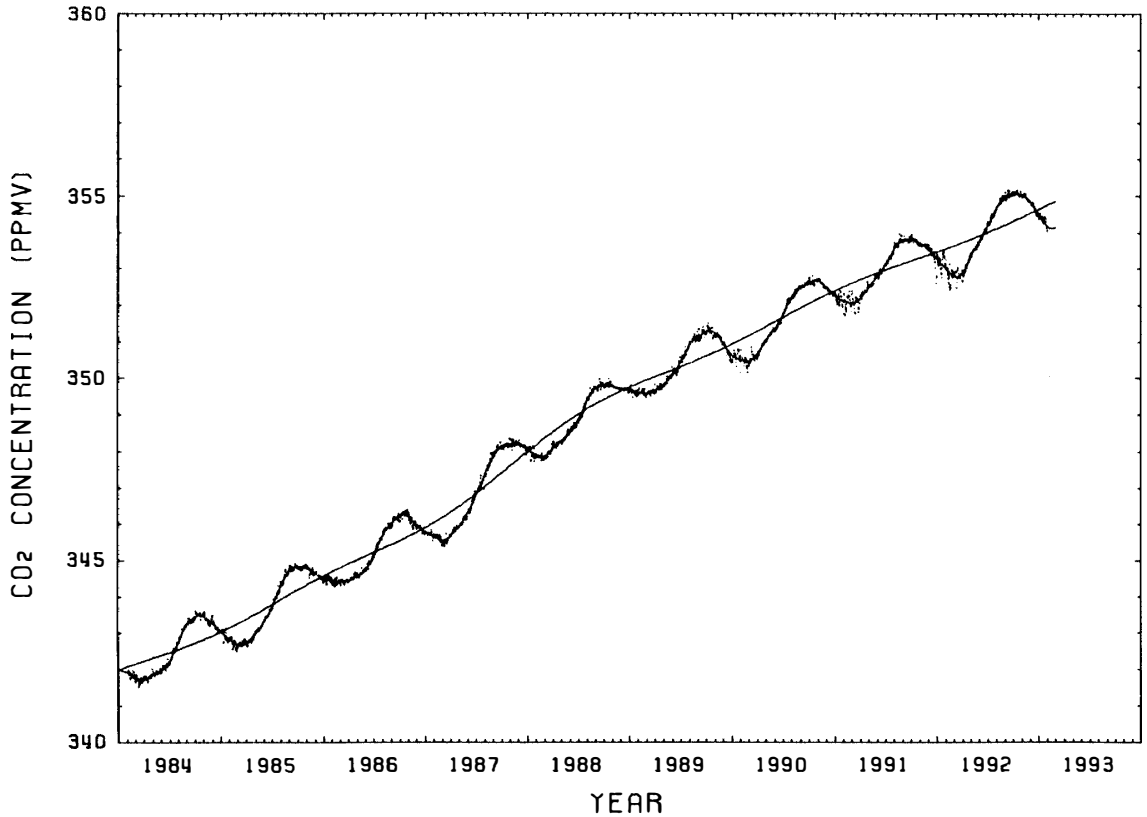

Fig. 1. Daily mean CO_2 concentrations at Syowa Station calculated from selected data. Solid lines denote the fitted curve of the data and the secular CO_2 trend.

cycle and irregular variations with shorter periods. In order to separate these components, a digital-filtering technique including Fourier harmonics, Reinsch-type cubic spline and Butterworth filter was used. The technique has been outlined elsewhere (*cf.* Nakazawa *et al.*, 1991b).

The average rate of annual CO_2 increase for the period from February 1984 to January 1993 is 1.54 ppmv yr^{-1}. The secular CO_2 trend is variable with time. The increase rate was especially large around the end of 1987; this is thought to be related to the 1986/1987 ENSO event. The average seasonal cycle of the CO_2 concentration reaches a minimum and maximum in late March and late September, respectively; its peak to peak amplitude is 1.18 ppmv. The amplitude of the observed seasonal CO_2 cycle is variable from year to year. The largest and smallest amplitudes appeared in 1992 and 1988, respectively, but there is no indication of a long-term increase of the amplitude. We found that the phase of the seasonal CO_2 cycle advances with time from the beginning of the systematic measurements; the dates when the seasonal CO_2 cycle reaches the maximum and minimum appeared earlier by about 20–30 days for the last 9 years, due probably to change in air transport. Irregular CO_2 variations with amplitude of about several tenths of ppmv and periods of a few weeks are also seen; this is primarily ascribed to exchange of different air masses in association with synoptic scale weather disturbances (Nakazawa *et al.*, 1991a). Irregular variations were enhanced, especially in the summers of 1990, 1991 and 1992.

2.2. *CH_4 concentration*

Measurement of the atmospheric CH_4 concentration was initiated at Syowa Station in February 1987. Grab sampling with subsequent laboratory analysis was also done from February 6, 1987 to February 16, 1988. The CH_4 concentration measured at the station was extremely stable, and no outlier due to local contamination was found throughout the year. Therefore, all the available data were used in this analysis. Daily mean values of the CH_4 concentration from continuous measurements and the CH_4 concentration obtained by grab sampling are shown in Fig. 2. In order to separate the secular trend, seasonal cycle and irregular variations of the CH_4 concentration, the digital-filtering technique described above was employed.

The average rate of annual CH_4 increase from February 1987 to January 1993 was 10.4 ppbv yr^{-1}. The rate of CH_4 increase was variable with time, showing significantly high values, especially in 1991. The enhancement of the increase rate was also observed between 1990 and 1991 by our aircraft measurements over Japan (our unpublished data). The average seasonal cycle of the CH_4 concentration shows minimum and maximum concentrations in early May and late September, respectively, and a peak to peak amplitude of 29.5 ppbv. The amplitude and phase of the observed seasonal CH_4 cycle have been almost the same for the last 6 years, which is different from the result for the CO_2 concentration; deviations of the observed seasonal cycle from the average cycle were within 5% for CH_4 and 15% for CO_2. This may suggest that the CH_4 concentration varies seasonally in phase in the southern hemisphere, while the

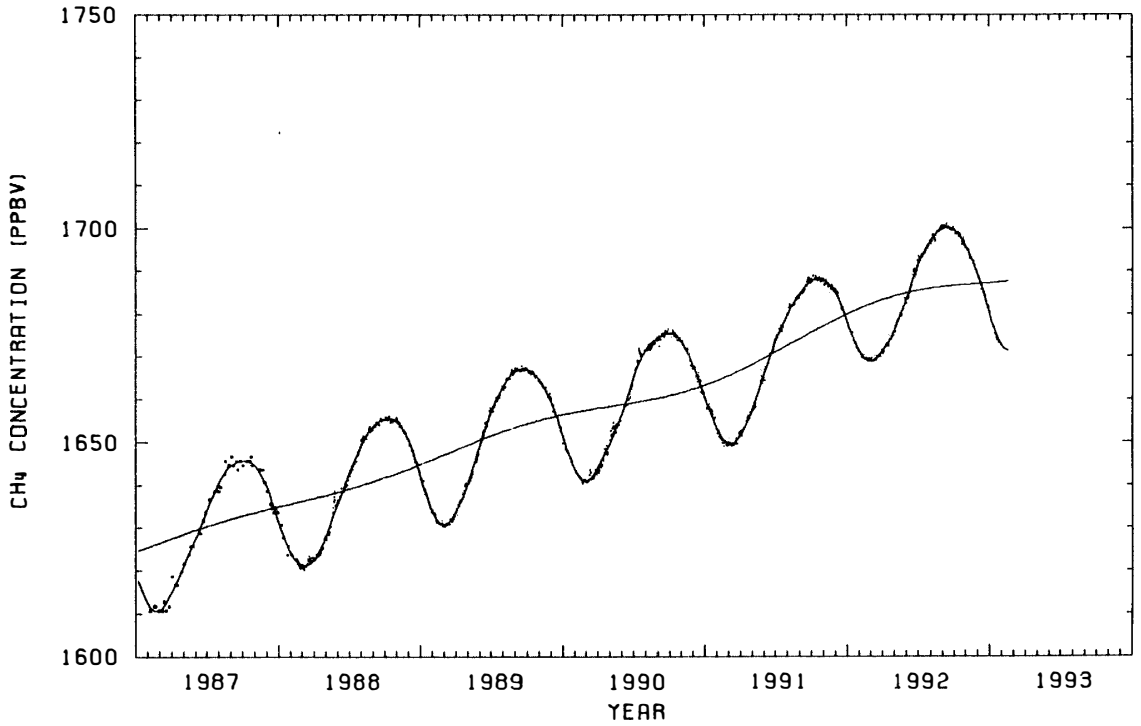

Fig. 2. Daily mean CH_4 concentrations at Syowa Station calculated from all available data. CH_4 concentrations obtained by grab sampling method are also plotted. Solid lines denote the fitted curve of the data and the secular CH_4 trend.

CO_2 concentration is spatially different, perhaps due to different seasonal CO_2 cycles in different regions.

2.3. *Relationship between secular trends of the CO_2 and CH_4 concentrations*

Figure 3 shows the increase rate obtained by differentiating the secular trends of the CO_2 and CH_4 concentrations. It is clearly seen in this figure that the rate of CO_2 increase is negatively correlated with that of CH_4 and that both rates oscillate with periods of 2.3 to 2.8 years. The increase rate of the CO_2 concentration was most enhanced in late 1987; the rate exceeded 2.2 ppmv yr^{-1}. Similar enhancements of the CO_2 increase are also seen in mid 1985, mid 1990 and late 1992. On the other hand, the increase rate of the CH_4 concentration reached high values of about 18 ppbv yr^{-1} in mid 1991. The rate of CH_4 increase also shows high values in early 1987 and early 1989. Taking account of the large negative correlation of the increase rate between CO_2 and CH_4, it may be suggested that global budgets of CO_2 and CH_4 are disturbed by some common cause such as climatic change. For a better understanding of the carbon cycle on the earth's surface, it is very important to elucidate its cause.

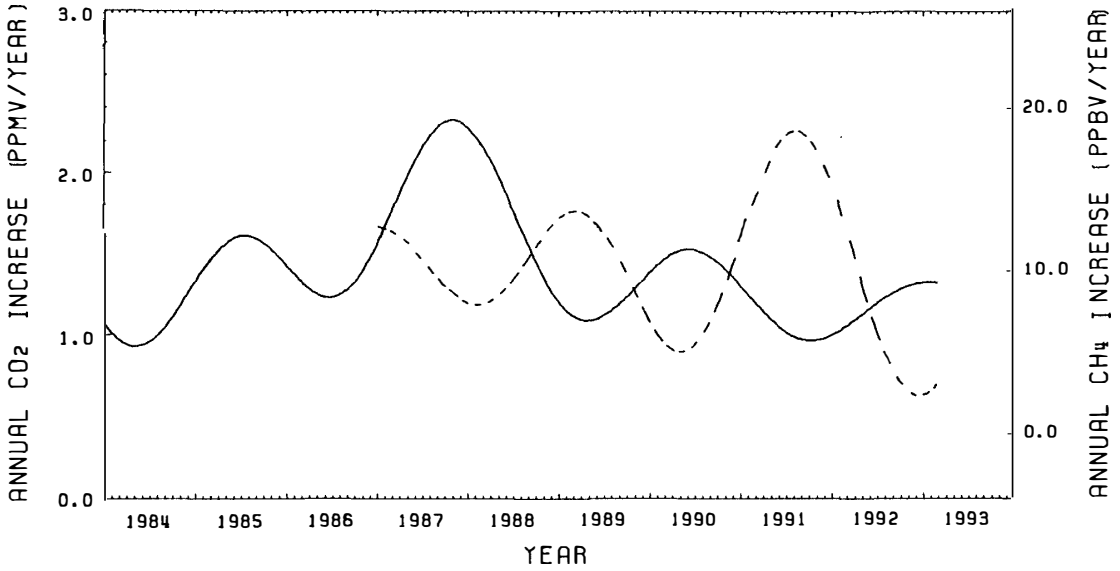

Fig. 3. Rates of annual increase for the CO_2 (solid curve) and CH_4 (dashed curve) concentrations at Syowa Station.

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(Received November 30, 1993; Revised manuscript received March 28, 1994)