## 4. METHANE (CH<sub>4</sub>)

## $\bullet: IN SITU \text{ STATION} \\ \Delta: FLASK \text{ STATION}$



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Methane (CH<sub>4</sub>) is the second most significant greenhouse gas in terms of its contribution to global warming. From 1000 to 1800 A.D., the concentration of CH<sub>4</sub> was about 700 ppb and varied within 40 ppb in synchronization with climatic variations (Etherridge *et al.*, 1998). The global average was estimated at 1730 ppb in 1997 (WMO, 1999a). The increase in the atmospheric CH<sub>4</sub> concentration over industrial and agricultural periods has brought about positive radiative forcing that amounts to about 30% of that for CO<sub>2</sub> (IPCC, 2001).

Atmospheric CH<sub>4</sub> is emitted mainly from anaerobic processes and fossil fuel exploitation, and removed by reaction with hydroxyl radicals (OH). Sources of CH<sub>4</sub> are both natural and anthropogenic, including natural wetlands, oceans, landfills, rice paddies, enteric fermentation, gas drilling and biomass burning. CH<sub>4</sub> sinks involve reaction with OH in the troposphere and the stratosphere, reaction with chlorine atoms and oxygen atoms in an excited state of O(<sup>1</sup>D) in the stratosphere and soils. However, uncertainties concerning the sources and sinks of CH<sub>4</sub> must be further reduced to improve estimates in the atmospheric CH<sub>4</sub> budget. Carbon isotopic studies should provide constraints for the budget.

At the top page of this chapter, operational observation sites on the ground that submitted  $CH_4$  concentration data to the WDCGG are shown on a map.

Global, hemispheric and zonal mean concentrations were provided based on data from 83 stations (points) that can be considered to provide background data.

Figure 4.1 shows the monthly mean concentrations and their deseasonalized long-term trend from 1984 to 2001 for the globe and both hemispheres. A long-term increase and seasonal variation are seen as for CO<sub>2</sub>. Amplitudes of the seasonal cycle are larger in the Northern Hemisphere than in the Southern Hemisphere. The annual mean concentration for the globe is 1782 ppb in 2001. Figure 4.2 shows the growth rates for the same areas as in Fig.4.1. Growth rates are generally lower in the 1990s than in the 1980s. Averaged global growth rates are 8 ppb/year for the whole period (1984-2001), 11 ppb/year for 1984-1990 and 5 ppb/year for 1991-2001. The decreased growth rates in the 1990s suggest that global CH<sub>4</sub> concentrations are approaching a steady state where removal of CH<sub>4</sub> is balanced with emission from CH<sub>4</sub> sources (Dlugokencky *et al.*, 1998 and Etherridge *et al.*, 1998). During the analysis period, global growth rates were at a maximum in June 1984 at 14 ppb/year and at a minimum in October 2000 at -1 ppb/year. Negative global growth rate in 2000 that suggest the global sink was stronger than the global sources was the first appearance during the analysis period. Reliability of the increasing feature in the latter half of 2001 may be inferior, because a guess value is used in this trend analysis near the end of the analysis period as explained in 2.4.

Figure 4.3 shows a time series of global CH<sub>4</sub> growth rates and their comparison with the global mean temperature anomaly of 1000hPa, which is calculated from NCEP reanalysis data and processed by a five-month running mean. Global CH<sub>4</sub> growth rates varied to correspond with global mean temperature anomalies especially in the 1990s. High temperature anomaly

brings about an increased CH<sub>4</sub> emission from wetlands and an increased removal by increased OH radicals (Bekki *et al.*, 1997). The relation between global growth rates and temperature anomaly shows that the former effect exceeds the latter globally. A study of the relationship between CH<sub>4</sub> concentrations in ice cores or firn layer and global temperature anomalies (Etherridge *et al.*, 1998) also suggests that a large growth rate for the CH<sub>4</sub> concentration may follow a high global mean temperature.

Figure 4.4 shows the monthly mean concentrations and their deseasonalized long-term trend from 1984 to 2001 for each of the 30-degree latitudinal zones. Deseasonalized long-term trend curves have the distinct feature of high concentration in northern high and mid-latitudes and low concentration in southern latitudes, with a larger gradient in the Northern Hemisphere than that in the Southern Hemisphere. According to the measurements of CH<sub>4</sub> concentrations in Antarctic and Greenland ice cores, inter-polar differences varied between 24 and 58 ppb from 1000 to 1800 A.D. (Etherridge *et al.*, 1998). At present, the difference between the northern and southern high latitudes is about 150 ppb. This increase is presumably due to increased emissions in the Northern Hemisphere, which are mainly from anthropogenic sources.

Figure 4.5 shows the analytical results of the deseasonalized long-term trend and growth rates. Growth rates clearly decreased from the 1980s to 1990s for all of the latitudinal zones. Large interannual variations overlay this trend. Growth rates were large in 1991 and small in 1992 and 1993 in general, and in 1992, the values were negative in northern high latitudes. The large increase in 1991 may have been caused by decreased OH radicals due to weakened UV radiation, an impact of the eruption of Mt. Pinatubo in 1991 (Dlugokencky *et al.*, 1996). The decrease in 1992 was probably caused by a decrease in fossil fuel production (Dlugokencky *et al.*, 1994) and biomass burning (Lowe *et al.*, 1997) along with an increase in OH radical concentration due to stratospheric ozone depletion after the eruption of Mt. Pinatubo in 1991 (Bekki *et al.*, 1994). Growth rates were large in 1998 for all of the latitudinal zones. Dlugokencky *et al.* (2001) suggested that the large growth rates in 1998 were attributed to the increased emissions from the northern high latitudinal zones and the tropical wetlands due to the high temperature and increased precipitation, and partly the influence of the biomass burning in the boreal area mainly in Siberia.

Figure 4.6 draws averaged seasonal cycles for each of the 30-degree latitudinal zones. Seasonal cycles are brought about mainly by reaction with OH, a major CH<sub>4</sub> sink in the atmosphere. The strength and timing of the emission from CH<sub>4</sub> sources such as wetland and biomass burning along with the transportation of CH<sub>4</sub> also affects the seasonal cycle. Amplitudes of the seasonal cycle are large in the Northern Hemisphere. Unlike CO<sub>2</sub>, amplitudes are large in the Southern Hemisphere except for low latitudes because OH radicals are distributed in both hemispheres, which brings about a similar seasonal variation. CH<sub>4</sub> concentrations are at a maximum in January or February in the Northern Hemisphere and in September in the Southern Hemisphere, except in low latitudes. They are at a minimum in July in the Northern Hemisphere and in February or March in the Southern Hemisphere, except in low latitudes. The timing of the maximum and minimum concentrations of CH<sub>4</sub> is consistent

with seasonal variation in OH concentration through photochemical reactions. Southern low latitudes have a distinct semi-annual component overlaying the annual component of the seasonal cycle. The secondary maximum occurs in boreal winter due to the large-scale transportation of CH<sub>4</sub> from the Northern Hemisphere. This phenomenon is seen at stations located in the western Indian Ocean, e.g. Mahe Island, Seychelles and the western and central equatorial Pacific, e.g. Cape Matatula, Samoa. However, it is not seen at Ascension Island in the equatorial Atlantic, where CH<sub>4</sub> is transported on a large scale from the Southern Hemisphere in boreal winter.