

## 過去25年間の成層圏CO<sub>2</sub>濃度のトレンドと年々変動 Trend and interannual variation of the stratospheric CO<sub>2</sub> in the past 25 years

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Systematic collections of stratospheric air samples have been carried out over Japan since 1985, using a balloon-borne cryogenic sampler. The stratospheric air samples have been collected almost once a year or two years at 11 assigned heights, ranging from the tropopause to 30 - 35 km. The air samples were analyzed for various gas concentrations, such as CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, and SF<sub>6</sub>, and their isotopes. Measurements of the stratospheric CO<sub>2</sub> concentration are one of the most promising methods to detect possible changes in the stratospheric circulation, because chemical loss and production are negligible in the stratosphere and its long-term trend in the troposphere is propagated into the stratosphere, with some time lag. Increasing trend of the CO<sub>2</sub> concentration was clearly found at heights above 20-25 km, where the CO<sub>2</sub> concentration becomes almost constant vertically. To clarify the difference of the secular CO<sub>2</sub> increases between the mid-stratosphere and the troposphere, the average values of the CO<sub>2</sub> concentration, calculated from the balloon data obtained at heights above 20-25 km, were compared with annual mean CO<sub>2</sub> concentrations at Mauna Loa (MLO) observed by NOAA/ESRL. The average increase rate of the CO<sub>2</sub> concentration in the mid-stratosphere, calculated by using a least-squares method, was 1.55(+/-0.03) ppmv/year. This value is significantly smaller than 1.73(+/-0.03) ppmv/year calculated for the same period for MLO data. Considering that the mid-stratospheric CO<sub>2</sub> concentration corresponds to the tropospheric values earlier by 4-5 years, the CO<sub>2</sub> increase rate in the stratosphere should be compared with the tropospheric values shifted by the same years. The average increase rate, thus calculated for the period 1981-2005, was 1.62(+/-0.03) for MLO data. This value is slightly smaller than those described above, due to interannual variations of CO<sub>2</sub> increase rate in the troposphere, but still larger than the stratospheric value. These facts imply that the concentration difference between the troposphere and mid-stratosphere gradually increased during the last 25 years. The interannual CO<sub>2</sub> variation in the mid-stratosphere was first discovered by our balloon measurements. The secular CO<sub>2</sub> increase in the mid-stratosphere is not monotonous, probably due to the propagation of interannual variations in tropospheric CO<sub>2</sub>, being accompanied by time delay. The CO<sub>2</sub> anomalies in the mid-stratosphere, calculated as deviations from the second order polynomial trend and then shifted by -4.5 years, are fairly correlated with those in the troposphere. Such a correlation is found especially in CO<sub>2</sub> anomalies observed in the troposphere for a few years after 1991, which is known as the Pinatubo anomaly. This result suggests that the stratospheric air age can be newly estimated from the phase shift of the interannual CO<sub>2</sub> variations.

キーワード: 二酸化炭素, 成層圏, 長期トレンド

Keywords: CO<sub>2</sub>, Stratosphere, Long-term trend