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Ten-year measurements of the tropospheric O2/N2 ratio over Japan and estimation of global CO2 budget

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The atmospheric O_2/N_2 ratio has been observed by several laboratories since the early 1990's to estimate terrestrial biospheric and oceanic CO_2 uptake. Most of those observations have been carried out at ground-fixed stations and ships. To examine height-dependent behavior of the O_2/N_2 ratio, as well as to estimate the global CO_2 budget, we initiated systematic measurements of the O_2/N_2 ratio at three different altitudes of the troposphere over Japan in May 1999 and continued them until the present, in addition to those at the ground-fixed station in Sendai, Japan.

Air samples were collected once per month at altitudes of 2 and 4 km over the Pacific Ocean about 50 km off the coast of Sendai (38˚N, 140˚E) using a chartered light aircraft, and at 8 km-the tropopause over the main island of Japan between Sendai and Fukuoka (34˚N, 130˚E) using commercial jet airliners. Air sampling was also made at the ground site in the suburbs of Sendai. The air samples were collected into Pyrex glass flasks with Viton O-ring seal at ambient pressures, and then analyzed for the O_2/N_2 ratio with a precision of $\pm$5.4 per meg using a mass spectrometer. The CO_2 concentration for each sample was determined by analyzing a simultaneously-collected pressurized air sample with a precision of $\pm$0.05 ppm using a NDIR analyzer. The results obtained from these measurements and analyses of their data are summarized as below:

The observed O_2/N_2 ratio shows a secular decrease, in contrast to secularly increasing CO_2 concentration. The O_2/N_2 ratio also shows a prominent seasonal cycle at all altitudes, which varies almost in opposite phase with that of the CO_2 concentration. The amplitude and phase of the seasonal O_2/N_2 ratio cycle are generally decreased and delayed, respectively, with increasing altitude; the seasonal maximum and minimum are found later by about two months, and the amplitude is reduced by 63% at 8 km-tropopause, compared to the seasonal cycle at the ground surface. Annual averages of the observed O_2/N_2 ratio and CO_2 concentration values generally increase and decrease, respectively, with increasing altitude. The average rate in change of the O_2/N_2 ratio relative to altitude between 2 km and 8 km-tropopause is found to be 2.85 per meg km^{-1}, while the corresponding rate for the CO_2 concentration is -0.30 ppm km^{-1}. It is also found that the O_2/N_2 ratio and CO_2 concentration are significantly affected by ENSO at all heights. By assuming that the results of the O_2/N_2 ratio and CO_2 concentration observed over Japan are representative of their global average secular trends, we estimate average terrestrial biospheric and oceanic CO_2 uptake over the observation period to be 1.1$\pm$0.8 and 2.8$\pm$0.7 GtC yr^{-1}, respectively.