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Comparison of -O2/CO2 ratio in pollution events observed at Hateruma with those of emission inventories and a simulation

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The National Institute for Environmental Studies (NIES) has been carrying out in-situ measurements of atmospheric CO2 mole fraction and O2/N2 ratio at Hateruma Island (HAT, 24 deg. N, 124 deg. E) since October 1993 and October 2006, respectively. An NDIR analyzer and a gas chromatograph equipped with a TCD have been used for CO2 and O2/N2, respectively. Synoptic scale pollution events with abrupt CO2 increases and O2 decreases were often observed when air masses were transported from the East Asian regions.

O2 and CO2 fluxes are tightly coupled in fossil fuel/biomass burning and in land biotic processes, respiration and photosynthesis. The estimated -O2/CO2 molar exchange ratios for fossil fuel burning vary depending on the types of fossil fuels: 1.17 +/- 0.03 for coal, 1.44 +/- 0.03 for liquid fuel and 1.95 +/- 0.04 for natural gas. Therefore, relative changes in the atmospheric O2 and CO2 concentrations for pollution events could be used to constrain compositions of fuel types at the pollution sources. For example, -O2/CO2 molar exchange ratios of fossil fuel burning and cement production for China, Japan and Korea from the CDIAC inventory in 2006 are 1.11, 1.37, and 1.31, respectively. The rather lower ratio for China is attributed to the fact that coal burning and cement production account for 73% and 10% of the total fossil CO2 emissions in recent years. These different -O2/CO2 molar exchange ratios for emissions from East Asian countries would likely appear in the observations at HAT.

In order to examine the influences of East Asian emissions on the O2 and CO2 observations at HAT, we calculate the individual -O2/CO2 changing ratio for each pollution event (total 123 events) obtained during October 2006 - December 2008. Then we categorize the air mass origins at pollution events as China, Korea, Japan and other using 3-day backward trajectories calculated by METEX (METeorological data Explorer). We find that the -O2/CO2 changing ratios for the events of China origin seem to be low in comparison with those for Japan and Korea origins. The observed average -O2/CO2 changing ratios and their standard errors are 1.12 +/- 0.02 for China, 1.40 +/- 0.09 for Japan, and 1.37 +/- 0.05 for Korea, which agree well with the above mentioned CDIAC inventory values.

In the above analysis, an air mass origin is assigned to be a single source region. However, actual transports are more complicated: the observed pollution events should be influenced to some extent from multiple source regions. In order to examine the influence of more realistic transport, we simulate the atmospheric O2 and CO2 changes at HAT with a Lagrangian particle dispersion model, FLEXPART. In this simulation, we take into account CO2 and O2 fluxes from fossil fuel burning and cement production (FF&C), terrestrial biosphere, and ocean. FF&C O2 flux is produced from the FF&C CO2 flux by scaling with -O2/CO2 molar exchange ratios based on the CDIAC national inventory. Note that the individual national -O2/CO2 ratios are used for China, Japan, and Korea while the average -O2/CO2 ratio is used for the other countries. O2 terrestrial

biosphere flux is derived from the CO2 terrestrial biosphere flux by scaling with the -O2/CO2 molar exchange ratio of 1.1.

The result confirms the most of CO2 and O2 changes in the pollution events at HAT are attributed to the FF&C emissions and the contribution from terrestrial biosphere is negligible. There is good agreement of the -O2/CO2 changing ratios between the observation and simulations for China, while the differences in the -O2/CO2 changing ratios between China and Japan/Korea, which are seen in the observations, disappear in the simulation. These results might be attributed to insufficient temporal and spatial resolution of meteorological data and/or fluxes used in the simulation.

Keywords: atmospheric oxygen, atmospheric carbon dioxide, transport, FLEXPART, Lagrangian particle dispersion model, O2/CO2 ratio