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波照間で観測される汚染イベント中-O₂/CO₂変動比と排出インベントリおよびモデル計算値に基づく-O₂/CO₂変動比との比較

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

峰島 知芳^{1*}, 久保 恵美², 遠嶋 康徳¹, 山岸 洋明¹, 古山祐治¹, 向井人史¹,
シャミールマクシュートフ¹, 北 和之²

Chika Minejima^{1*}, Megumi Kubo², Yasunori Tohjima¹, Hiroaki Yamagishi¹, Yuji Koyama¹,
Hitoshi Mukai¹, Shamil Maksyutov¹, Kazuyuki Kita²

¹国立環境研究所, ²茨城大学

¹NIES, ²Ibaraki University

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 (METeoro logical 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.

キーワード:大気中酸素,大気中二酸化炭素,大気輸送, FLEXPART,拡散モデル, O₂/CO₂変動比

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