論文の内容の要旨

論文題目：**Studies on ozone photochemistry over the western Pacific in winter and spring**

(西太平洋域における冬季・春季のオゾン光化学過程の研究)

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Investigations of photochemical production and transport of ozone (O_3) over the Asian continent and the western Pacific are essential for the understandings of regional and global O_3 distributions because increase of anthropogenic emissions from East Asian region is considered to lead to regional and global O_3 enhancements. However, comprehensive measurements in winter and late spring are still very limited. The purpose of this work is to reveal the O_3 photochemistry over these regions in winter and spring using the datasets obtained from the aircraft measurements and three-dimensional (3-D) chemical transport model. By combining these results, we could have a better understanding of photochemistry of O_3 during the transport from the Asian continent (source region) to the western Pacific (receptor region).

O_3, its precursors, and photolysis frequencies were observed during the Pacific Exploration of Asian Continental Emission (PEACE) aircraft campaigns conducted by

the Japan Aerospace Exploration Agency (JAXA) over the western Pacific in January (PEACE-A) and April-May (PEACE-B) 2002. These measurements have provided datasets that, in combination with the NASA Transport and Chemical Evolution over the Pacific (TRACE-P) data taken over the same region in March 2001, enable studies of O_3 photochemistry from winter to late spring. The species which were not observed such as OH and HO_2 and photochemical O_3 formation and destruction rates ($F(O_3)/D(O_3)$) were calculated using the photochemical box model constrained with observed parameters.

These key parameters revealed vertical distributions and seasonal changes of photochemical O_3 productions. As shown in Figure 1, the net O_3 production rate ($P(O_3)$), given by $F(O_3)$-$D(O_3)$, was largely positive in the boundary layer (0-3 km) at 30-45 °N (1.5-4 ppbv day^{-1}) in January, due mainly to high NO and low H_2O values. Positive $P(O_3)$ continued from January to the end of March, demonstrating that the western Pacific is an important O_3 source region during these seasons. $P(O_3)$ nearly ceased by late-April/May due to the decrease in NO and the increase in H_2O. In the latitude range of 20-30 °N, $P(O_3)$ in the boundary layer was positive in January and turned negative by March. The earlier transition was mainly due to lower NO and higher H_2O concentrations, combined with weaker transport and higher temperatures than those at 30-45 °N. In the middle troposphere (3-6 km), O_3 production hardly occurred. It is because of lowest NO_x in this altitude. The upper troposphere (6-12 km) has been shown to be a region of net O_3 formation throughout most of the year due to relatively high NO and low H_2O. The observational study illustrates that a decrease in the net O_3 production rate at 20-45 °N latitude from winter to late spring is explained systematically by the increases in $J(O^1D)$, H_2O, OH, and HO_2 and the decrease in NO.

Variations of O_3 over East Asia and the western Pacific from winter to spring were investigated in terms of photochemistry and transport using the 3-D CTM GEOS–CHEM. For the validation of the GEOS-CHEM results for January and April 2002. they were compared with data obtained by the PEACE aircraft observations made at the same periods. Ozone, its precursors, and $P(O_3)$ calculated by GEOS–CHEM generally agreed well with the observed and box model values for this region.

The NO_x in the boundary layer (0-2.1 km) was highest (several ppbv) over industrial regions of the Asian continent, two orders of magnitude higher than that over the western Pacific. Thus, $F(O_3)$ was highest over the Asian continent and decreased with distance from the continent to the western Pacific. The maximum HO_x radicals occurred 10-20 °N and decreased with latitude. The spatial distribution of $D(O_3)$ was similar to that of HO_x. As a result, $P(O_3)$ were positive over the Asian continent but negative over the western Pacific (Figure 2), indicating that O_3 was persistently produced over the Asian continent. The increase in $P(O_3)$ with season over the Asian continent was mainly caused by the increase in photochemical activity. In contrast, it significantly decreased with season due to weakened transport of NO_x in April.

Monthly mean O_3 in the boundary layer are also shown in Figure 2. In January, the O_3 mixing ratios were highest in the downstream regions of the Asian outflow. In April. the O_3 mixing ratios were highest over regions around the Japan Sea. The mean flow patterns suggest that these high O_3 were due to accumulation of O_3 during transport of continental air in the high $P(O_3)$ regions. Anthropogenic NO_x emitted from the Asian continental region caused 1-3 ppbv (January) and 3-10 ppbv (April) of O_3 increases in the downwind regions.

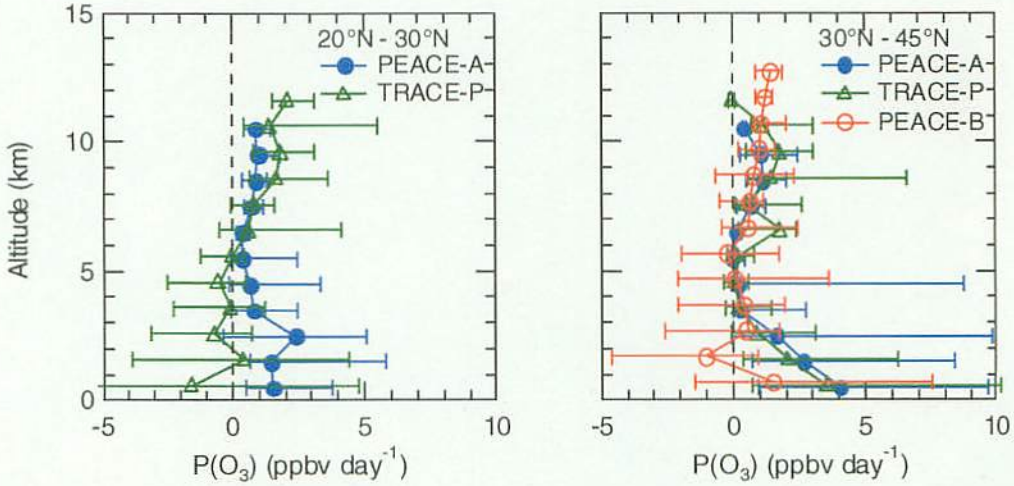

Figure 1. Vertical profiles of the net formation ($P(O_3)$) rate at 20°N-30°N and 30°N-45°N in units of ppbv day^{-1}.

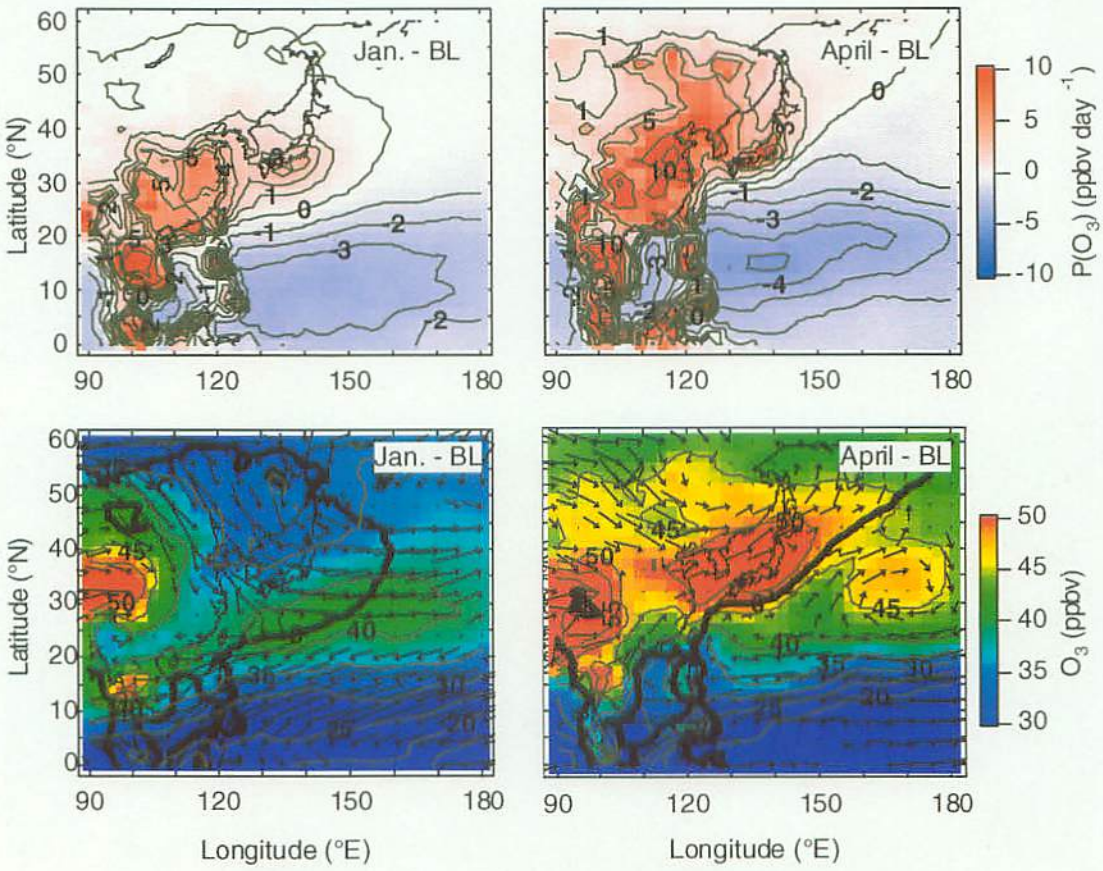

Figure 2. Monthly mean $P(O_3)$ (top) and O_3 (bottom) in January (left) and April (right) in the boundary layer simulated by GEOS-CHEM. Mean wind fields and the line where $P(O_3)$ is zero (thick line) are also displayed in bottom panels.