

Observations of O₃, CO, CO₂ and CH₄ concentrations at Happo and estimations of the source by chemical transport model

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A large increase in tropospheric O₃ concentrations was observed during spring for the period from 1998 to 2006 at Mt. Happo Observatory (36.7°N, 137.8°E, 1840 m asl), which is one of the Acid Deposition Monitoring Network in East Asia (EANET) stations (Tanimoto, 2009). The increase in the springtime O₃ reproduced by a regional chemistry-transport model incorporating the updated anthropogenic emissions inventory in East Asia can only explain about half of the observed O₃ increase (Tanimoto et al., 2009). On the other hand, previous source-receptor analysis by a global chemistry-transport model indicated that the contributions of local and regional sources can vary, depending on individual episodes (Wild et al., 2004). For better understanding of the discrepancies between the model prediction and observational evidence, it is necessary to better characterize air masses at Mt. Happo by simultaneous measurements of additional tracers like CO, CO₂, and CH₄.

Starting in July 2013, we have made continuous measurements of CO, CO₂ and CH₄ as well as O₃ at Mt. Happo. We found that the O₃ levels at Mt. Happo increased until 2007, and then the increase slowed down and now decreased to the same level as in 1990s. The CO concentrations observed for the period of 2013-2014 was found to be lower than those in 1990s. In particular, the current CO levels were 50 ppbv lower than in 1990s during summer. In total, 44 events associated with O₃ enhancement were identified for the period from July 2013 to August 2014. Correlations of CO with CO₂ and CH₄ with CO₂ were used to identify possible source regions for individual episodes. Emission ratios ($\Delta\text{CO}/\Delta\text{CO}_2$ and $\Delta\text{CH}_4/\Delta\text{CO}_2$) were calculated in each event, and compared with the ratios estimated by the regional emission inventory in Asia (REAS; Kurokawa et al., 2013). There is a discrepancy between two emission ratios. The $\Delta\text{CO}/\Delta\text{CO}_2$ indicated that most events except summer were affected by the emissions from East Asia. On the other hand, the $\Delta\text{CH}_4/\Delta\text{CO}_2$ indicated that most of events were caused by the emissions from Japan and Korea. It is difficult to identify the source regions by the ratios only.

We compared the observations with the model calculations by CMAQ v4.7.1, a regional chemistry-transport model, with the horizontal and vertical resolutions of 80 km and 37 layers, respectively. The model calculations well reproduced variability and seasonal changes of CO. For O₃, although the model prediction was higher than the observed, in particular during the summer season, the model reproduced the O₃ enhancement events reasonably well. The source regions inferred by the model will be discussed in the presentation.

Keywords: ozone, carbon monoxide, methane, carbon dioxide