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Investigation of NO_x emissions and NO_x-related chemistry in East Asia using CMAQ-predicted and GOME-derived NO_2 columns

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Abstract. In this study, NO_2 columns from the US EPA Models-3/CMAQ model simulations carried out using the 2001 ACE-ASIA (Asia Pacific Regional Aerosol Characterization Experiment) emission inventory over East Asia were compared with the GOME-derived NO_2 columns. There were large discrepancies between the CMAQ-predicted and GOME-derived NO_2 columns in the fall and winter seasons. In particular, while the CMAQ-predicted NO_2 columns produced larger values than the GOME-derived NO_2 columns over South Korea for all four seasons, the CMAQ-predicted NO_2 columns produced smaller values than the GOME-derived NO_2 columns over North China for all seasons with the exception of summer (summer anomaly). It is believed that there might be some error in the NO_x emission estimates as well as uncertainty in the NO_x chemical loss rates over North China and South Korea. Regarding the latter, this study further focused on the biogenic VOC (BVOC) emissions that were strongly coupled with NO_x chemistry during summer in East Asia. This study also investigated whether the CMAQ-modeled NO_2/NO_x ratios with the possibly overestimated isoprene emissions were higher than those with reduced isoprene emissions. Although changes in both the NO_x chemical loss rates and NO_2/NO_x ratios from CMAQ-modeling with the different isoprene emissions affected the CMAQ-modeled NO_2 levels, the effects were found to be limited, mainly due to the low absolute levels of NO_2 in summer. Seasonal variations of the NO_x emission fluxes over East Asia were further investigated by a set of sensitivity runs of the CMAQ model. Although the results still exhibited the summer anomaly possibly due to the uncertainties in both NO_x-related chemistry in the CMAQ model and the GOME measurements, it is believed that consideration of both the seasonal variations in NO_x emissions and the correct BVOC emissions in East Asia are critical. Overall, it is estimated that the NO_x emissions are underestimated by ~57.3% in North China and overestimated by ~46.1%

in South Korea over an entire year. In order to confirm the uncertainty in NO_x emissions, the NO_x emissions over South Korea and China were further investigated using the ACE-ASIA, REAS (Regional Emission inventory in ASia), and CAPSS (Clean Air Policy Support System) emission inventories. The comparison between the CMAQ-calculated and GOME-derived NO_2 columns indicated that both the ACE-ASIA and REAS inventories have some uncertainty in NO_x emissions over North China and South Korea, which can also lead to some errors in modeling the formation of ozone and secondary aerosols in South Korea and North China.

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