Seasonal variation of formation, loss, and transport of HNO3 and particulate nitrate over the Tokyo Metropolitan Area

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Simulations of particulate nitrate (NO_3^-) by three dimensional chemical transport models (3-D CTM) are generally difficult mainly because there are large uncertainties in the production and removal rates of NO_3^- and nitric acid (HNO_3). Time-resolved (1 min - 1 h) measurements of HNO_3, NO_3^-, and other important gas and aerosol species were conducted at an urban site in Tokyo in winter and summer of 2004. Intensive measurements were also conducted at a suburban (downwind) site during the summer of 2004. In this study, a 3-D CTM (CMAQ) is used to simulate the production, transport, and removal processes of HNO_3 and NO_3^- during the measurement period. The model reproduces observed seasonal-diurnal variations of HNO_3 and NO_3^- concentrations. The sensitivities of the total nitrate (TNO_3 = HNO_3 + NO_3^-) concentrations on the N_2O_5 uptake coefficient on aerosol particles, g_{N2O5}, and HNO_3 dry deposition velocity, $v_{d,HNO3}$, are assessed. This analysis indicates that g_{N2O5} and $v_{d,HNO3}$ are critical parameters in predicting TNO_3 concentrations. The loss rate of total reactive nitrogen (NO_y) was dominated by the removal of TNO_3. We evaluated the removal processes of TNO_3 by using remaining fraction of NO_y ($R(NO_y)$), which is defined as the dNO_y/dCO ratio normalized by the NO_x/CO emission ratio. The base case simulation largely overestimates $R(NO_y)$. We have found that the model well reproduces the observed $R(NO_y)$ when the dry deposition velocity of TNO_3 is increased by a factor of 5. This result suggests a need for improving the removal scheme of TNO_3 in current CTMs.