別紙1

論文の内容の要旨

Long-range Transport and Source-receptor Relationships of Acidifying Substances in East Asia
(東アジアにおける酸性化物質の長距離輸送と排出—受容関係)

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The impact of global and regional air pollution on climate and the environment is a new focus of atmospheric science. Sulfur dioxide (SO_2) and nitrogen oxides ($NO_x = NO + NO_2$), mainly emitted from fuel combustion, can be transformed in the atmosphere to aerosol-bound sulfate and nitrate. Aerosols, which have lifetimes of approximately one to two weeks, can be transported over thousands of kilometers resulting in transboundary pollution problems. Nitrogen oxides play a key part in the photochemically induced catalytic production of ozone (O_3), which results in summer smog and has increased levels of tropospheric ozone globally. NO_x concentrations in many industrialized countries are expected to decrease, but rapid economic development has the potential to increase significantly NO_x emissions in parts of Asia. Chemical Transport Models (CTM) are playing increasingly important roles in the studies of long-range transport, chemical transformations and scavenging of trace gases and aerosols in the troposphere. Regional exchange of sulfur compounds in East Asia has been widely studied mainly using simple Lagrangian models during the recent decade. The three-dimensional (3-D) Eulerian models are able to handle more accurately the transport of pollutants in the free troposphere above the boundary layer as contrast to the simple Lagrangian models. However, regional exchange of reactive nitrogen ($NO_y = NO + NO_2 + NO_3 + 2N_2O_5 + HONO + HNO_4 + HNO_3$ + aerosol nitrate + PAN(s) + other organic nitrates) in East Asia have not yet until now, to date, been identified quantitatively by a complex 3-D Eulerian model.

This study has aimed at establishing region-to-grid source-receptor relationships (SRRs) for sulfur and reactive nitrogen deposition in East Asia to support possible integrated assessments of transboundary pollution. A multi-scale long-range transport modeling system has been set up, developed, and evaluated in East Asia for this purpose. The state-of-the-science 3-D Eulerian-type Community Multiscale Air Quality model (CMAQ) is applied in East Asia with horizontal resolutions of 27-km and 81-km. The meteorological fields for the two domains are generated by Mesoscale Model version 5 (MM5) using the NCEP/NCAR global meteorological reanalysis datasets in 2001 for initial and boundary conditions. An emission processing model is newly developed to import and transform various inventory data by seasonal allocation, chemical speciation, vertical assignment (only for large point sources), and horizontal interpolation. To suppress the pronounced inaccuracy of time-invariant lateral-fixed boundary conditions, the seasonal outputs of the global Model for Ozone and Related Chemical Tracers (MOZART) are processed by a newly developed global/regional interface to derive the boundary conditions for the 81-km grid domain of CMAQ. The MOZART-derived boundary conditions are able to account for the global influence to regional air quality in East Asia caused by the long-lived pollutants such as ozone, carbon monoxide, peroxyacetyl nitrate (PAN), and aerosols.

In order to characterize model performance and to build confidence in its use, observational data used in this study include ground-based monitoring from the Acid Deposition Monitoring Network in East Asia (EANET) and satellite measurements from the Global Ozone Monitoring Experiment (GOME) spectrometer. A variety of results and analysis are presented to evaluate how well the current emission estimates and the multi-scale modeling system can represent the observed features of gases and aerosols over a wide range of meteorological conditions and geographical areas.

Firstly, the spatial and temporal variations (from seasonal to diurnal) of ambient concentrations of primary gases SO_2 and NO_x are analyzed and evaluated. The model successfully reproduces the magnitudes, daily, and diurnal variations of SO_2 mixing ratios at most of the EANET sites especially those with 27-km grid spacing. Main uncertainty of SO_2 predictions is caused by the representation of model topography and sub-grid variation of emissions in urban areas, which may not be resolved in the coarse 81-km grid resolution. The magnitudes of NO_x surface concentrations at the EANET sites in central and southern Japan are slightly underpredicted. Through comparison with the GOME retrieval, the model is shown to be able to capture major spatial and seasonal variations of tropospheric NO_2 column abundance in East Asia. Regarding the magnitudes of NO_2 columns, however, the model largely (by a factor of 2) underpredicts GOME retrieval over the industrialized area of Central Eastern China in winter, suggesting that anthropogenic NO_x emissions over this region, especially domestic heating sources, are likely underestimated in the current emission inventory. In addition, the model also underpredicts GOME retrieval over the remote western China and Mongolia in summer. This is likely caused by the underestimation of soil-biogenic NO emission from the grassland/scrubland/desert over these regions during the wet season.

Secondly, the model's ability to reproduce the photochemical production of O_3 is evaluated by the analysis of photochemistry sensitivity with the CBIV and SAPRC99 mechanism. Analysis of seasonal cycle of surface O_3 suggests that the use of MOZART-derived boundary conditions improves the O_3 prediction during the winter season when the inflow from Europe across Eurasia is significant. The exchange between continental polluted air mass and clean marine air mass results in the summer minimum of ozone over the south of 30°N in the study region. The summer-time O_3 production in central eastern China and central Japan is highly sensitive to the chemical mechanisms applied in CMAQ. Both 27-km and 81-km simulations of the CBIV mechanism well reproduce the observed seasonal cycle of surface ozone at most sites. Although the SAPRC99 mechanism provides a more-detailed representation of hydrocarbon classification, the 81-km simulation largely overpredicts the observed summer-time O_3 especially on low O_3 days near central Japan.

Thirdly, precipitation and the chemical components in rainwater are analyzed. The model generally reproduces ionic concentrations in rainwater and annual wet deposition loads of sulfate and ammonium. Except the uncertainty in NO_x emissions, CMAQ has a tendency to underestimate the production of nitrate aerosol. More integrated observation of reactive nitrogen gases and aerosols are needed to evaluate the thermodynamic aerosol module of CMAQ.

Finally, the complex 3-D Eulerian model CMAQ and related components newly developed in this study are applied, for the first time, to quantify region-to-grid source-receptor relationships (SRRs) both for sulfur and reactive nitrogen in East Asia. Investigation of model responses to emission changes suggest that reductions of ammonia emissions have significant nonlinear effects on the deposition of sulfur and reactive nitrogen as a result of non-linearity in the atmospheric chemistries of sulfate-nitrate-ammonia system and the deposition process. A source region attribution methodology is proposed based on the analysis of non-linear effects. Sensitivity simulations were conducted where emissions of SO_2, NO_x, and its primary particles from a specific source region were reduced by 25%. The difference between the base and sensitivity simulations was multiplied by a factor of four, and then defined as the contribution from that source region. Source attributions are carried out for eighteen designated source regions as well as international shipping and volcanoes within the study domain.

Region-to-region quantitative SRRs provide an important connection between emissions and fates of pollutants over different time and spatial scales. The transboundary influences exhibit strong seasonal variation and are generally maximum during the dry seasons reflecting the Asian monsoon circulation. Long-range transport from central eastern China contributes a significant percentage (>20%) of anthropogenic sulfur deposition as well as reactive nitrogen deposition throughout Asia. At the same time, northwestern China receives approximately 35% of sulfur load and 45% of nitrogen load due to foreign emissions mainly from the Indian subcontinent. Volcanic sources including emissions from Miyakejima volcano in 2001 contribute approximately 50% of total sulfur deposition in Japan. Sulfur inflows from regions outside the study domain, which is attributed from the MOZART-derived boundary conditions, are pronounced (10~40%) over different parts of Asia. Compared with previous studies using simple Lagrangian models, the results derived in this study show more effects from long-range transport. The relationships are more realistic including the global influences and internal interactions among different parts of China. Region-to-region source-receptor relationships of acid deposition provide quantitative information for environmental negotiation, cost allocation, and possible international initiatives to mitigate global air pollution and climate change.