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ESTIMATION OF NITROGEN AND SULFUR DRY DEPOSITION OVER THE WATERSHED OF LAKE PALDANG IN KOREA

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1. INTRODUCTION

In Korea, more than 90% of drinking water is produced from surface waters including rivers and reservoirs (Park, 1999). This means that water supply systems are vulnerable to contamination by various pollutants and at risk due to accidents. Recently, it has been suggested that intake water for producing drinking water is affected by air pollutants. Any pollutants in the air can fall and affect the water quality (USEPA, 1997). Among them, sulfur and nitrogen compounds acidify lakes and streams. Acidification also appears to mobilize toxic metals such as aluminum and mercury. Excess nitrogen can cause eutrophication (over enrichment of nutrients) in nitrogen-sensitive waters such as bays and estuaries, and increase nitrate concentrations in drinking water supplies. It is known that in major rivers of the northeastern U.S., nitrate concentrations have risen three- to ten-fold since the early 1900s, and the evidence suggests a similar trend in many European rivers (Vitousek, 1997).

The greater Seoul area (GSA) – which includes Seoul proper and its neighboring satellite cities – accounts for about 40% of Korea's population but less than 5% of its total land. Seoul, which has an area of 606 km^2, is crowded with 2.3 million cars and 10 million people. Lake Paldang is a main resource of drinking water for 20 million people in the greater Seoul area. In this study, dry deposition amounts of nitrogen and sulfur were estimated for three typical days in each season over the watershed of Lake Paldang.

Models-3/CMAQ (USEPA Models-3/Community Multi-scale Air Quality) and MM5 (PSU/NCAR Mesoscale Modeling System) were used to predict air quality and meteorology, respectively. Both gas and particulate species were considered.

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2. MODELING

2.1 *Target Domain and Period*

The modeling domain was 480 km × 348 km centering on watershed of Lake Paldang. The grid size was 12 km × 12 km and the number of grids was 40 in the west-east direction and 29 in the south-north direction. The number of layers in the vertical direction was six to the height of 100 hPa. The Lambert Conformal projection was used for the map.

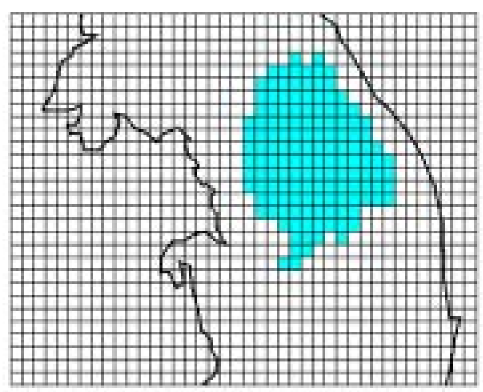

Fig. 1 Modeling domain. Colored area is the water shed of Lake Paldang.

Three consecutive days were selected as episode days in each season for the year of 1997. There was basically no precipitation for six days including the previous three days for spin up; air temperature and wind characteristics were close to those of a normal year (KMA, 1991). Out of four seasons, wind speed was the highest in spring and the lowest in fall.

2.2 *Input Data*

The emission data with resolution of 6′ × 6′ grid size were taken from the University of Iowa prepared for ACE-Asia (Asian Pacific Regional Aerosol Characterization Experiment). Especially, a detailed emission data with resolution of 1 km × 1 km were used for the GSA and Kangwondo area that have high emission density. Biogenic emissions of isoprene and terpene were taken from GEIA (2003). Point sources were excluded in this study because of a large uncertainty of the emission data. Seasonal variation of the emission was considered only for the biogenic emission. Because traffic volume is the main factor affecting the diurnal variation of emission in the GSA, 130% of the hourly average emission amounts for 07:00-19:00 LST and 70% of the hourly average emission amounts for the remaining 12 hours was assumed. The default data set provided with MM5 was used as the landuse data. Sample data provided for the test run of Models-3 (probably clean air conditions) were used for initial and boundary conditions. The first 63 hours were devoted to a spin-up period in order to minimize the influence of assumed homogeneous initial conditions. The NCEP/NCAR (National Center for Environmental Prediction/National Center for Atmospheric Research) reanalysis data of six-hour intervals were used for initial and boundary conditions of MM5.

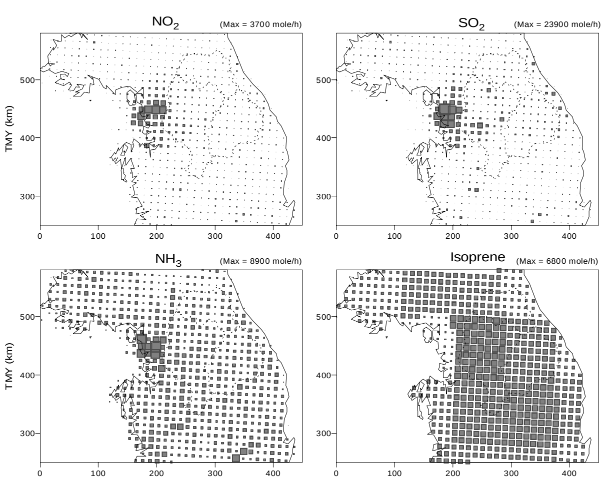

Fig. 2 Distribution of pollutant emissions.

3. RESULTS AND DISCUSSION

3.1 *Concentration Variations*

The predicted concentrations of NO_2 and SO_2 are compared with observed ones in Seoul (Fig. 3). The observed ones are average of measurements of 20 monitoring stations in Seoul, and the predicted ones are average of concentration at four grids over the Seoul area. NO_2 and SO_2 generally vary in the similar range with the observed one, despite of somewhat underestimation. Underestimation is probably caused from faster wind speed of MM5 than the observed one.

Average diurnal variations of gaseous and particulate pollutants are compared in each season over the watershed of Lake Paldang. NO_2, SO_2 and NH_3 concentrations are high at nighttime and low at daytime because of variations of mixing height and wind speed. Nitrate and ammonium show the highest concentration in winter along with NO_2. HNO_3 and sulfate are highest in fall because pollutants generated in GSA are transport to the watershed of Lake Paldang.

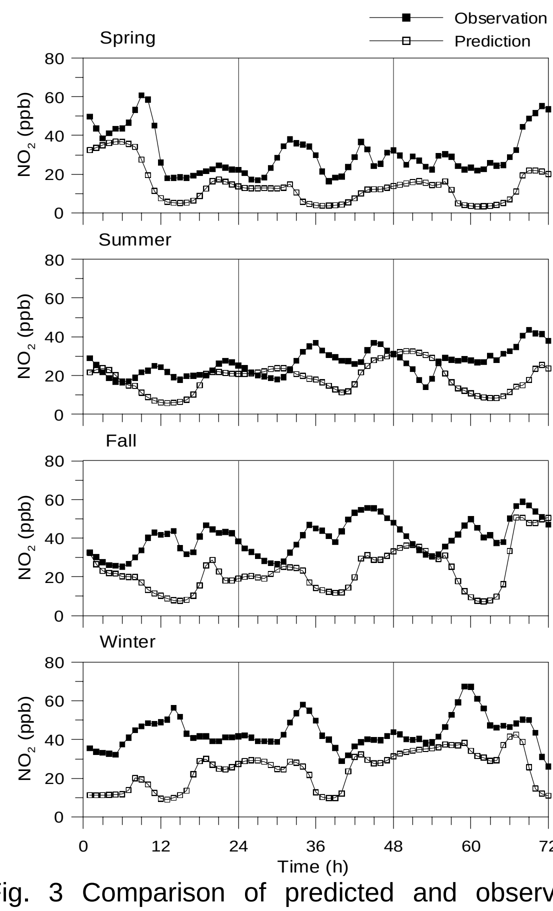

Fig. 3 Comparison of predicted and observed concentrations in Seoul, Korea.

3.2 *Deposition Variations*

Fig. 4 shows the average diurnal variation of dry deposition velocity over the watershed of Lake Paldang. Deposition velocity of nitric acid is the highest. The velocities are higher at daytime and rather constant at night. It is interpreted that high deposition velocities at daytime are due to relatively high wind speeds and atmospheric instability. This indicates that the effect of variation in the aerodynamic resistance is dominant over those of quasi-laminar sublayer resistance and surface resistance at daytime. Especially, dry deposition velocity of nitric acid is the highest in spring when wind speed is the highest. This is because the surface resistance of nitric acid is particularly low; and, as a result, aerodynamic and quasi-laminar sublayer resistances become a main factor in determining the dry deposition velocity.

Deposition velocities of NO_2, SO_2 and HNO_3 are comparable to other values of references (Finlayson-Pitts and Pitts, 1986; Brook et al., 1999; Park et al., 2000).

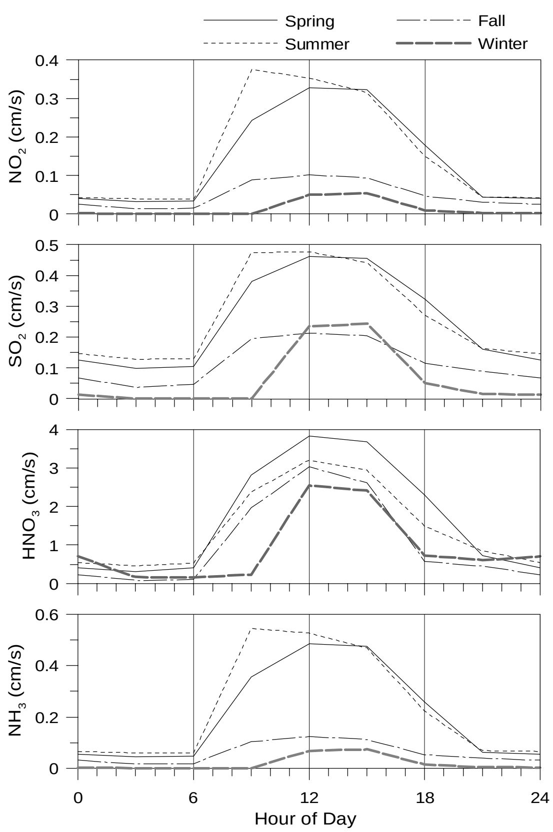

Fig. 4 Average diurnal variation of dry deposition velocity over the watershed of Lake Paldang.

Deposition fluxes are concentrations multiplied by deposition velocities. The fluxes are generally high in the season when the concentrations are high. NO_2 and SO_2 deposition fluxes are high in spring and summer because of high deposition velocities. HNO_3 deposition flux is large at daytime because of both high deposition velocity and high concentration. NO_3^- and NH_4^+ deposition fluxes are the highest in winter when concentrations are high. SO_4^{2-} deposition flux is the highest in fall when concentration is high. SO_4^{2-} deposition flux is also high in the afternoon. Average ranges of dry deposition flux over the watershed of Lake Paldang are NO_2 0.0-0.03 kg/km^2/hr, HNO_3 0.1-0.4 kg/km^2/hr, NO_3^- 0.0-0.03 kg/km^2/hr, NH_4^+ 0.0-03 kg/km^2/hr, NH_3 0.0-0.009 kg/km^2/hr, SO_2 0.0-0.05 kg/km^2/hr, SO_4^{2-} 0.0-0.008 kg/km^2/hr.

3.3 *Deposition Estimation*

The deposition amount was calculated at each grid point from the deposition flux for three episode days in each season (Fig. 5). Total nitrogen dry deposition is generally large in GSA; it is more dispersed to the east of GSA in spring, to the west-east direction of GSA (east-preferred) in fall, and to the southeast in winter. Therefore, dry deposition amount over the watershed of Lake Paldang is the highest in fall and the lowest in winter.

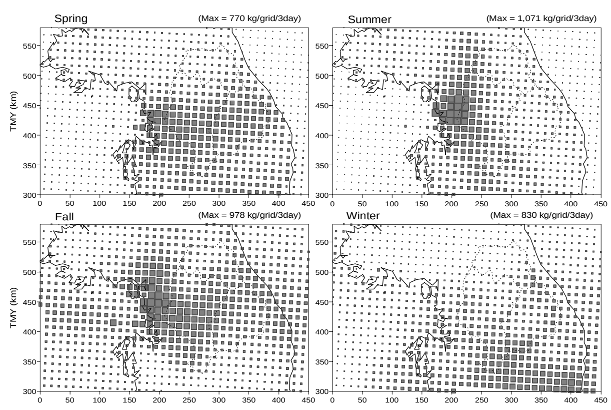

Fig. 5 Dry deposition distribution of total nitrogen for each season for 3 days.

The deposition distribution of oxidized nitrogen and reduced nitrogen is very similar with that of total nitrogen. However, deposition amount of oxidized nitrogen is much larger than that of reduced nitrogen; most of nitrogen is deposited in the oxidized form.

The deposition distribution of total sulfur is similar with that of total nitrogen in spring and summer, but it is higher over the sea in fall and winter. This is because SO_2 dry deposition velocity is higher over the sea because of its solubility.

Table 1 shows total amounts of nitrogen and sulfur deposition for three episode days in each season. Nitrogen deposition is large in fall due to

Table 1. Nitrogen and sulfur deposition on nitrogen and sulfur base, respectively, for three episode days in each season (unit: tons). The number in the parentheses represents the percent fraction.

	Spring	Summer	Fall	Winter
NO_2	5,175 (14.6)	4,924 (21.2)	1,907 (4.6)	1,536 (7.8)
NO	0.3 (0.0)	0.2 (0.0)	0.2 (0.0)	0.2 (0.0)
HNO_3	26,740 (75.5)	14,276 (61.3)	37,270 (90.2)	13,179 (67.0)
NH_3	2,277 (6.5)	3,643 (15.7)	963 (2.3)	333 (1.7)
NO_3^-	323 (0.9)	1.1 (0.0)	152 (0.4)	2,108 (10.7)
NH_4^+	894 (2.5)	429 (1.8)	1,037 (2.5)	2,513 (12.8)
Total N	35,409 (100)	23,273 (100)	41,329 (100)	19,669 (100)
SO_2	15,270 (95.7)	14,817 (96.4)	7,876 (86.7)	9,894 (94.0)
H_2SO_4	48.1 (0.3)	18.4 (0.1)	10.6 (0.1)	7.8 (0.1)
SO_4^{2-}	645 (4.0)	541 (3.5)	1,195 (13.2)	622 (5.9)
Total S	15,963 (100)	15,376 (100)	9,081 (100)	10,523 (100)

large deposition of nitric acid. Nitric acid is the most contributing pollutant to the total nitrogen deposition. Contribution of NO_2 deposition is 21% in summer, 15% in spring and less than 10% in other season. Contribution of NH_3 deposition is 16% in summer. Contribution of particulate nitrogen deposition is high in winter, about 25%. Sulfur deposition is large in spring and summer. Most contributing pollutant to the sulfur deposition is SO_2. Contribution of particulate sulfur deposition is high in fall.

It can be estimated that annual depositions of nitrogen and sulfur over the watershed of Lake Paldang are 3,645 tons and 1,553 tons, respectively. Annual emissions of nitrogen and sulfur are 8,971 tons and 5,301 tons, respectively. Therefore, the annual dry deposition amount is 41% for the nitrogen and 29% for the sulfur compared with the emission over the watershed of Lake Paldang.

4. ACKNOWLEDGEMENTS

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