C-4.1.2 Studies on Control and Evaluation of Acid Rain Precursors in East Asia

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### Abstract

As Japan is located at the eastern extreme of East Asia, there are concerns about the transboundary transport of acid rain pollutants carried from continental China by the westerlies from autumn through winter and into spring. The exceptionally fast recent development of China's industrial regions suggests that the impacts of acid rain will continue to grow. We selected 10 major industrial areas mainly along China's eastern coastal region, and simulated the state of transboundary pollution through wet acidic fallout caused by sulfur oxides from each of these areas. The receptor sites in Japan were 47 national or local government-operated monitoring stations from Hokkaido to Kyushu that are capable of measuring sulfuric acid ions.

The simulation time frame was the two months of October and November 1993, and the simulation used the parameters of each month.

SO<sub>2</sub> was calculated by assuming a yearly 5% increase from the emissions reported for the 10 areas in 1987, and the results were used as 1993 SO<sub>2</sub> emissions. Simulation results showed that emissions from the coastal industrial zones in eastern and southeastern China have a large impact on Japan's Kyushu region, while those from China's northeast affect Japan's Kanto and Tohoku regions. Actual results of measurements in some locations, however, fiffered from those anticipated, indicating that more study of the accurary of measurements of SO<sub>2</sub> emissions and sulfuric acid ions, meteorological date, reaction speeds in the atmosphere, the applicability of the model used, and other items is needed.

Keywords: China; westerlies; acidic fallout; transport model; simulation

### 1. Introduction

East Asia has the third-highest emissions of the acid rain-causing substances  $SO_2$  and  $NO_X$  after the European and North American continents. China, the country that will most likely directly affect both South Korea and Japan with acid rain, already has  $SO_2$  emissions of about 2O million tons annually, and there are concerns about future impacts owing to China's economic development from here on.

This research used data on hand to simulate the extent to which Japan is currently affected by the areas of China with the most rapid industrial development. It is hoped that the results might underpin future measures to deal eith this problem.

- 2. Conditions for Transport Model and Parameters
- 2-1 General Description of the Model
- (1)Basic Formula for Dispersion Model

The basic dispersion model formula incorporates terms for formation and consumption into Ficks' dispersion equation. The model's coefficients were chosen with reference to the RAINS model, an acid rain projection model developed in Europe. Pollutants dispersion is shown by the following differential equation:

#### Where:

C is the mass of substance per unit volume

V is the wind speed vector

 $K_D$  is the dispersion coefficient

E is the substance formation term

R is the substance removal term

K is the term for formation and consumption caused by chemical reactions.

To solve for differences the model used iterative wind difference calculus for the advection term. As it is known that the term for horizontal dispersion is one to two orders smaller than the term for horizontal advection, the model avoided becoming a discrete difference, and the equation was solved by the Krank-Nicholson method. The dispersion coefficient was estimated, fot stable atmospheric conditions, with O'Brien's equation, and for unstable conditions with Cauthey and Palmer's equation.

## (2) Formation and Consumption

The two comounds  $SO_2$  and  $SO_4^{2-}$  were subjected to analysis for advection and dispersion, and the model took into consideration the transition from  $SO_2$  to  $SO_4^{2-}$ .

$$\frac{dC_{s02}}{dt} = -(\frac{-}{-} + k_x + k_{w1}) \cdot C_{s02} + (1 - \alpha - \beta) - \cdots (2)$$

$$\frac{dC_{S04}}{----} = (-- + k_{W2}) \cdot C_{S04} + k_{x} \cdot C_{S02} + \beta - \cdots$$

$$V$$

### Where:

 $C_{S02}$ ,  $C_{S04}$  are the atmospheric concentrations of  $SO_2$  and  $SO_4^{2-}(gS/m^3)$ 

 $V_1$ ,  $V_2$  are the dry deposition velocities of  $SO_2$  and  $SO_4^{2-}(m/sec)$ 

h<sub>4</sub> is the height at which dry deposition occurs(m)

 $K_X$  is the rate of transition from  $SO_2$  to  $SO_4^{2}$  (1/sec)

 $K_{W1}$ ,  $K_{W2}$  are the removal rates by rain for  $SO_2$  and  $SO_4^{2-}(1-sec)$ 

 $\alpha$  is the percentage deposited locally near the source(-)(assumed to be 0.15)

 $\beta$  is the percentage assumed to be directly formed as sulfates(-)(assumed to be 0.05)

Q is the rate of  $SO_2$  formation for each mesh cell per unit time(gS/sec) V is the volume of a mesh cell with a  $source(m^3)$ 

The lowest layer for  $h_4$  was set at 150m.  $\alpha$  and  $\beta$  were set at 0.15 and 0.05, respectively, which are the values used in the RAINS model.

# (1)Dry Deposition Velocity

Velocity was set at  $0.01_{\text{Inw15}}\text{m/s}$  for  $\text{SO}_4^{2-}$ , while the following formulas, in reference to the RAINS model, were used for  $\text{SO}_2$ :

$$al \cdot Vdl + (1 - al) \cdot Vdz \cdot tan(0.5(\pi/2-\phi):terrestrial)$$

$$Vd(1m) = \cdots \cdots \cdots \cdots (5)$$

$$0.008 : marine$$

#### Where:

Vd(lm) is dry deposition velocity at a height of lm (m/s)

al is a constant(0.5)

Vd1 is 0.002 from 0400-2200 hrs(daytime), and 0.004 from 2200-0400 hrs (nighttime)(m/s)

Vdz is the dry deposition velocity of  $SO_2$  that is dependent on latitude (0.008m/s)

 $\phi$  is the latitude of the location for which calculations are made (degrees)

 $\kappa$  is the Karman constant (0.4)

u (10m) is windspeed at 10m above the ground(m/s)

ZO is the roughness parameter(m)(marine: [illegible]; terrestrial: 0.25)

(2) Transition Rate from  $SO_2$  to  $SO_4^2$ 

The following equation was used for the transition rate k from  $SO_2$  to  $SO_4^{\,2}$ 

$$k_t = k_{to} + k_{ti} \cdot \sin(2\pi - \frac{\tau}{2}) \cdots (6)$$

### Where:

 $k_{to}$  is the constant term for the transition rate k from  $SO_2$  to  $SO_4^{2-}$  (3.0  $\times$   $10^{-3}$  S<sup>-1</sup>)

 $k_{t+}$  is the variation of the transition rate from SO<sub>2</sub> to SO<sub>4</sub>2-(2.0×10<sup>-3</sup> S<sup>-1</sup>)  $\tau$  is the month for which caluculations are made

### (3) Wet Deposition Rate

Rates at which rain removes  $SO_2$  and  $SO_4^{2-}$   $(k_{w1}$  and  $k_{w2})$  were estimated in the following manner.

## Where:

 $w_1$  is the SO<sup>2</sup> rainout rate (5.0×10<sup>5</sup>)

 $_{\rm w2}$  is the  ${\rm SO_4^{\,2}}^{-}$  rainout rate  $(5.6\times10^5)$ 

P is the surface precipitation amount per unit time(m/s)

h is the height of the upper border of the layer where precipitation  $begins(\mathbf{m})$ 

 $\nabla z$  is the thickness of the layer for which caluculations are made(m)

The model set the height at which precipitation begins at the upper border of the third layer  $(1.3 \, \text{km})$ , while it was set at the upper border of the second layer  $(550 \, \text{m})$  when elevation is  $1.3 \, \text{km}$  or higher.

# (3) Estimationg Acid Fallout

The Following equations were used to determine acid fallout by wet and dry deposition.

$$d_{d} = v_{1} \cdot t \cdot C_{S02} + v_{2} \cdot t \cdot C_{S04} \cdot \cdots \cdot (9)$$

$$d_{w} = (k_{w1} \cdot t \cdot C_{S02} + k_{w2} \cdot t \cdot C_{S04}) \cdot h \cdot \cdots \cdot (10)$$

$$d_{t} = d_{d} + d_{w} \cdot \cdots \cdot (11)$$

# Where:

- $d_d$  is the dry deposition per unit area in t seconds( $gS/m^2$ )
- $d_w$  is the wet deposition per unit area in t seconds( $gS/m^2$ )
- $d_t$  is the fallout per unit area in t seconds( $gS/m^2$ )

# 2-2 SOX Emissions on Major Areas of China

Emissions of the ten regions around Beijing, Shanghai, Chongqing, Tianjin, Shenzhen, Luoyang, Benxi, Nanjing, Hangzhou, and Shenyang were estimated by starting with the 1987 SO<sub>x</sub> emissions for Asia as studied under the Environment Agency's Global environment Research Support Fund and the Science and Technology Agency's "Asian Energy Use and the Global Environment," and then multiplying these results by each country's rate of increase up to 1993. Table 1 shows the 1993 emissions for areas extending 40-80 km from each city center.

## 2-3 Conditions Used in Calculations

# (1)Gridding

Domains were divided horizontally by a 40 km grid, and vertically into six layers with boundaries at heights of 150, 550, 1300, 3000, and 5000m from the ground.

## (2) Meteorological Data

The model used the wind speed component, atmospheric pressre, and rainfall intensity for October and November 1993 predicted by the European Center for Medium-range Weather Forecasting, as well as the precipitation from AMeDAS data.

Table -1 SO2 emissions in major cities of China

(unit:  $10^3 t/y$ )

city	emission
Beijing	513
Shanghai	1, 434
Chongqing	685
Tianjan	809
Shenzhen	290
Luoyang	297
Benxi	396
Nanjing	610
Hangzhou	290
Shenyang	215
China	28, 035
Korea	1, 764

## 3. Simulation Results

# 3-1 Locations of Major Areas

Fig. 1 shows the locations of the 10 areas in China, and Fig. 2 shows the acid rain evaluation locations.

## 3-2 Comparison of Simulation Results with Measurements

(1)Comparison of Simulation Results with Measurements by National Government Monitoring Stations

For government monitoring stations and other monitoring stations, which were selected according to the criteria listed below, we determined the correlation coefficients, regression coefficients, and the like for calculated and measured values, and used them to examine the simulation's applicability.

# Selection Criteria

- In Kyushu, stations affected by volcanoes were excluded.
- Large differences (50% or more) between the measured and calculated precipitation amounts were excluded as localized effects.
- Obviously localized effects in urban areas and other places were excluded.

Results from the selected monitoring stations show that the correlation coefficients are 0.5 and 0.8 for October and November, respectively. The correlation for November was better than that for October.

## (2) Impacts from Major Areas of China

Table 2 and 3 show the contributions by amount of wet fallout from the major areas of China to the evaluation locations.

In October the greatest impact of wet fallout from China overall was on Tsuruga City at  $0.15g/m^2$ , followed by Shizuoka City at  $0.12g/m^2$ .

The largest of the contributions to fallout in Japan from individual areas of China was the  $0.03 \mathrm{g/m^2}$  at Tsuruga City, followed by  $0.02 \mathrm{g/m^2}$  at Niigata City, both from Shanghai. Contributions from Beijing, Chongqing, and Shenzhen were found to be small.

The largest of the November wet fallout contributions from the various areas of China was  $0.03 \mathrm{g/m^2}$  at Matsue City from Tianjin, followed by  $0.02\,\mu\,\mathrm{g/m^2}$  at Tsuruga City from Shanghai. Just as in October, the effects from Beijing, Chongqing, and Shenzhen were small.

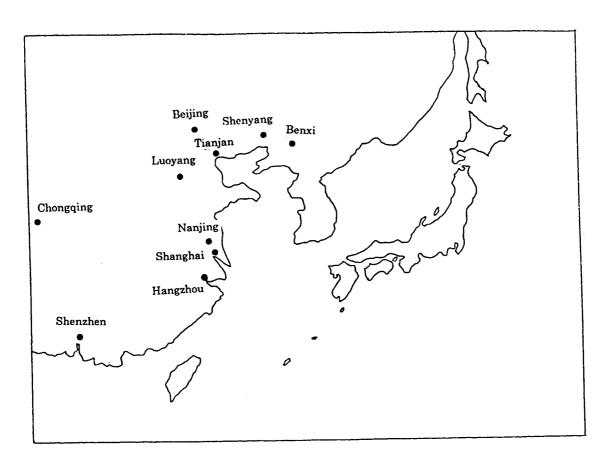


Fig-1 The locations of the 10 major cities of China

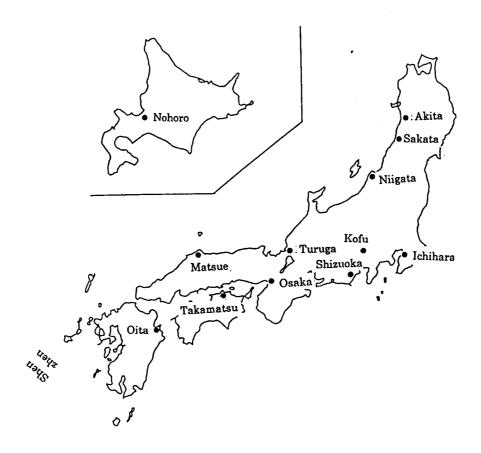


Fig-2 Selected monitoring sites for acid deposition evaluation in Japan

Table -2 The contributions by amount of wet fallout from the major cities of China to the selected sites of Japan (Oct. 1993)

receptor site	amount of wet fallout in October (g/m²)												
	All Areas	Bei jing	Shang hai	Chong qing	Tian jan	Shen zhen	Luo yang	Benxi	Nan jing	Hang zhou	Shen yang	China	Korea
Nohoro	0. 1107	0. 0001	0. 0009	0. 0001	0. 0058	0.0000	0. 0003	0. 0042	0. 0046	0.0000	0. 0021	0. 0538	0. 0121
Akita	0. 1799	0.0004	0. 0057	0. 0006	0. 0059	0. 0000	0.0020	0. 0028	0. 0030	0.0006	0.0016	0. 0945	0. 0347
Sakata	0. 0995	0.0004	0. 0028	0.0006	0. 0003	0.0000	0.0027	0.0001	0.0007	0. 0003	0.0001	0. 0491	0. 0491
Niigata	0. 2712	0.0001	0. 0223	0. 0006	0.0004	0.0000	0. 0007	0.0001	0. 0025	0.0017	0. 0002	0. 0885	0. 0462
Ichihara	0. 2394	0.0000	0. 0002	0. 0003	0. 0020	0.0000	0.0012	0.0011	0.0006	0.0000	0.0012	0. 0767	0. 0387
Kofu	0. 0876	0.0000	0. 0001	0. 0001	0. 0008	0.0000	0.0002	0.0005	0.0001	0.0000	0.0006	0.0413	0. 0329
Shizuoka	0. 2325	0.0000	0. 0001	0. 0005	0. 0055	0.0000	0.0007	0. 0058	0.0021	0.0000	0.0040	0. 1168	0. 0397
Turuga	0. 1956	0.0000	0. 0283	0. 0007	0.0048	0. 0002	0.0018	0.0061	0.0006	0. 0079	0.0027	0. 1492	0. 0158
Osaka	0. 1511	0.0007	0.0001	0.0006	0.0037	0.0000	0.0003	0.0134	0.0002	0.0000	0.0112	0. 0977	0. 0246
Takamatsu	0. 1828	0. 0026	0.0012	0. 0001	0. 0099	0.0000	0.0004	0.0041	0.0003	0.0003	0.0071	0. 0730	0. 0787
Matsue	0. 2012	0. 0001	0.0010	0. 0013	0.0017	0.0000	0. 0004	0. 0001	0.0020	0.0001	0.0003	0. 0561	0. 1199
Oita	0. 1507	0. 0001	0. 0002	0. 0002	0. 0009	0. 0000	0. 0026	0.0000	0.0016	0. 0000	0. 0000	0. 0605	0. 0185

Table -3 The contributions by amount of wet fallout from the major cities of China to the selected sites of Japan (Nov. 1993)

receptor site	amount of wet fallout in November (g/m²)										
	All Areas	Bei jing	Shang hai	Chong qing	Tian jan	Shen zhen	Luo yang	Benxi	Nan jing	Hang zhou	
Nohoro	0, 1279	0, 0019	0.0000	0.0013	0.0031	0.0000	0. 0010	0. 0050	0. 0002	0.000	
Akita	0. 2720	0. 0032	0.0003	0.0010	0.0088	0.0000	0.0044	0.0088	0. 0015	0.000	
Sakata	0. 4125	0. 0012	0.0019	0.0006	0.0134	0.0000	0. 0153	0.0105	0. 0016	0.000	
Niigata	0. 3035	0. 0007	0.0045	0.0018	0.0014	0.0000	0. 0024	0. 0009	0. 0031	0.000	
Ichihara	0. 1279	0. 0016	0.0000	0.0000	0.0027	0.0000	0. 0002	0.0016	0.0003	0.000	
Kofu	0. 0189	0. 0005	0.0000	0.0000	0.0017	0.0000	0. 0001	0.0002	0.0000	0.000	
Shizuoka	0. 1736	0.0010	0.0000	0.0000	0.0035	0.0000	0.0020	0. 0029	0.0010	0.000	
	0. 2591	0. 0052	0. 0240	0.0013	0.0029	0.0000	0.0004	0. 0107	0.0111	0. 002	
Turuga	0. 0880	0. 0002	0.0006	0.0003	0.0027	0. 0000	0. 0001	0.0001	0.0012	0.000	
Osaka	0. 5970	0.0010	0.0040	0.0027	0. 0291	0.0000	0. 0031	0.0007	0.0110	0.000	
Takamatsu	0. 0820	0. 0000	0.0002	0.0000	0.0004	0.0000	0.0011	0.0002	0.0107	0.000	
Matsue Oita	0. 1923	0. 0002	0.0008	0.0001	0. 0007	0. 0000	0. 0063	0.0000	0. 0127	0.00	

## 4. Discussion

We selected 10 areas primarily in China's eastern coastal industrial zone and performed a computer simulation on the impacts of wet acidic fallout on various locations in Japan. The simulation used data from meteorological observations for October and November 1993, and calculated the amounts transported to Japan on the basis of  $SO_2$  emissions amounts estmated for the above 10 areas.

As the simulation used meteorological observation values for only two months, it is possible that the results do not express the average autumnal weather conditions of East Asia. Further, emissions were calculated at a 5% annual increase from the estimated 1987 values. Because some people have expressed the opinion that those values are underestimated, one cannot expect to attain highly accurate figures for emissions based on these values. Nevertheless, this is perhaps the first attempt to assess the impacts of specific industrial areas in China on specific areas in Japan, and as such we believe it is quite valuable apart from the absolute accuracy of the results.

In future it will be necessary to increase the number of simulations and endeavor to improve the precision of simulations by means including: seeking meteorological data for times of average westerlies; making the maximum possible improvements in emissions data; improving the accuracy of receptor measurements; more precisely determining the atmospheric reaction process in order to provide models with highly accurate reaction coefficients for East Asia; and improving transport models.

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