C-1.2.2 Study on the Long-range Transport of Pollutants Related with Acid Precipitation

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Abstract To clarify the long-range transport process of pollutants related with acid precipitation in the east Asia, the long-range transport model has been developed. The transport model was consisted from two submodels, one was meteorological model and the other was dispersion model. The Lagrangian Particle was adopted in the dispersion model, and the random-walk method was used for the calculation of vertical diffusion process. The Monte Carlo calculation was adopted for dry deposition process. For wet deposition process, only below cloud scavenging was considered. The transformation process from sulfur dioxide to sulfate was also included in the model.

After making some validation of the model, the numerical simulation was performed for the transport process of sulfur oxides in east Asia all the year around 1985. The calculation was carried out by excepting the domestic emissions for evaluating the contribution on acid deposition in Japan from the emission in vicinal countries. The annual dry and wet depositions of sulfur oxides at several areas in Japan were evaluated.

The result of numerical simulation showed that the annual wet deposition of sulfur oxides in Japan originated in the emission in vicinal countries was 0.1-0.3 g/m^2. The calculated wet deposition of sulfur oxides was one order lower than observed wet deposition of sulfate.

Meanwhile, to grasp the vertical transport of sub-grid scale originated in topography, the field observation on vertical transport of aerosol was performed by using transportable and aircraft loading lidars. The observational results were reflected in the model.

Key Words Acid rain, Long-range Transport, Numerical model, wet deposition

1. Introduction

Recently, the extensive energy has been consumed in east Asia due to rapid progress of industrialization, in consequence, the air pollutants giving influence to environment has been emitted in the atmosphere. It is entertained apprehensions about the deposition of acidic substances to become serious in east Asia. The main objective of this study is to develop of numerical model on long-range transport incorporating dry and wet deposition components for evaluation of the extent of the acid deposition. Since the numerous transport models including wet deposition have been developed up to the present (Venkatram et al., 1982; Liu and Stewart, 1982), most of these models adopted finely observed meteorological data such as Winds field, precipitation etc.. As the most part of east Asian region is occupied by the ocean, there are not many available meteorological data observed continuously. The numerical model to predict the meteorological variables for the transport model was used.

2. The Brief Description of the Long-range Transport Model

The long-range transport model was consisted from two submodels, one was meteorological model to predict meteorological variables such as pressure, winds, temperature and humidity fields etc., and the other was dispersion model incorporating dry and wet depositions and transformation components. The constitution and flow chart of the long-range transport model is shown in Fig. 1.

2.1 Meteorological Model

The meteorological model adopted in the long-range transport model was the fine-mesh limited area model (FLM) which had been used operationally for the numerical weather forecast at Japan Meteorological Agency (JMA) for Asian region. The FLM was improved to be able to combine with dispersion model. The important points of improvement were additional vertical layer and physical process in the surface and planetary layers. The bulk method in the surface boundary layer and K-theory in the planetary boundary layer were substituted for the similarity theory and turbulent closure model of level-2 (Mellor and Yamada, 1974), respectively. The FLM adopted σ coordinate system.The global objective analysis (GANL) of JMA was used for initial and boundary conditions of the meteorological model. The integration was continued for each 36-hours by giving boundary condition at 12-hours intervals after integration started, namely, integration was begun from 12Z, and it was continued until 24Z of the next day. But the predicted meteorological variables for the portion of former 12-hours did not be avail because of the spinning up of the model. Only later 24-hours data were available. The cyclic integrations were carried out to get the meteorological variables for long period.

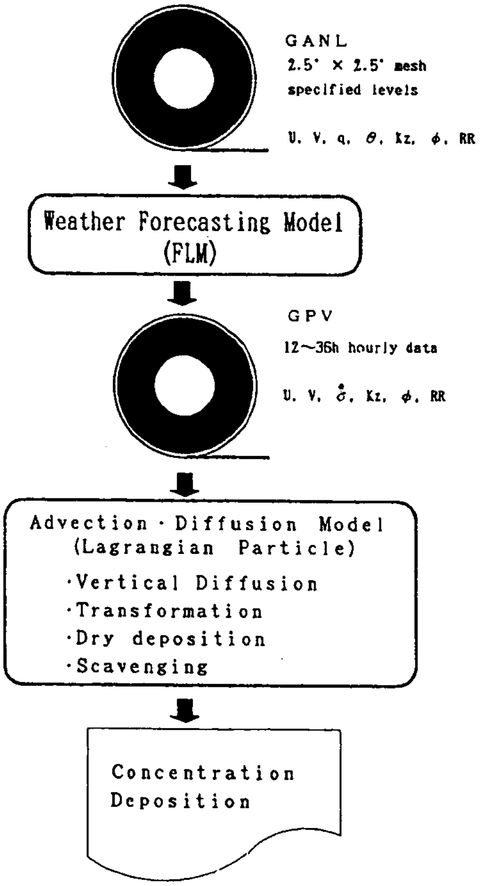

Fig. 1 Constitution of the long-range transport model.

Though the precipitation played important role on the acid deposition, the validation on the precipitation of the model was performed statistically by the use of AMeDAS data. And the determination of the threshold value on precipitation was made.

2.2 Dispersion Model

The Lagrangian particle is adopted in dispersion model, in which the movements of particles were calculated by 3 dimensional wind velocities provided by meteorological model. Furthermore, the random walk method was used to calculate the vertical turbulent diffusion, it gave additional random movements on the particles. The additional movements were randomly given by vertical velocity corresponding to vertical turbulent diffusivity.

For the vertical diffusivity, K_z, which was derived from meteorological model under the assumption as follows;

$$K_m=K_h=K_q=K_z=l^2\phi \qquad (1)$$

where, K_m, K_h, K_q are vertical diffusivity for momentum, heat and water vapor respectively, and l is mixing length, ϕ is defined as

$$\phi=[(\frac{\partial V}{\partial z})^2-\frac{g}{\theta}\frac{\partial \theta}{\partial Z}]^{\frac{1}{2}} \qquad (2)$$

here, $V^2=u^2+v^2$, θ is potential temperature.

The random walk method gave a much higher resolution than a grid method having the same grid size as that of the FLM. If the numbers of particle in an averaging area is small, the statistical error is large. The vertical coordinate of a particle at level z and time t is z(t), the position of particle after Δt is extrapolated by the equation;

$$z(t+\Delta t)=z(t)+\dot{\sigma}\Delta t\pm\sqrt{2\Delta t K_z} \qquad (3)$$

where plus or minus of the second term of right hand side is selected randomly for each particle and each time step. If the vertical diffusivity is constant and number of particles after a long enough time integration will be equal to an analytical solution of the diffusion equation.

Meanwhile, when vertical diffusivity depends on height z, however, an artificial vertical flux appears even though the vertical gradient of concentration is zero. Even if the time step is very small, this error will not becomes small. To avoid this error, Diehl et al., (1982) added a correction term to equation (3) so that the time extrapolation gives an exact flux when K_z is linear function of height. However, when K_z depends on height, so that the computation error here tends to be large. In this study, following scheme is used for time integration of the vertical diffusion. First, an estimated position of particle at t+Δt is made;

$$z^*=z(t)+\dot{\sigma}\Delta t\pm\sqrt{2\Delta t K_z} \qquad (4)$$

Then, the height of particle at t+Δt is estimated by using z^* as follow;

$$z(t+\Delta t)=z(t)+\dot{\sigma}\Delta t\pm\sqrt{2\Delta t K_z(z^*)} \qquad (5)$$

In using this scheme, the error will become small if Δt is sufficiently small.
Horizontal turbulent diffusion id neglected since this effect appears to be much smaller than the indirect horizontal diffusion caused by vertical wind shear. On the other hand, the

vertical turbulent diffusion is not negligible, since it is very important of pollutant at least during the first few days, and the major horizontal advection height of the pollutant is also important. If the travel time of the pollutant is enough long, i.e., much longer than the typical of a disturbance due to cyclone, vertical diffusion may becomes important than the vertical The Monte Carlo calculations were used for dry and wet depositions processes. vertical diffusion may becomes less important than the vertical advection due to disturbance. The horizontal advection of pollutant is depends on wind direction varying slightly with height.

Table 1 The dry deposition velocity and scavenging coefficient for each species.

	Species	Land	Water
Dry deposition velocity(cm/s)	SO_2	0.51	0.32
	SO_4^{2-}	0.21	0.02
Scavenging coefficient(1/s)	$S0_2$	$3x10^{-5}$	
	$S0_4^{2-}$	$1x10^{-4}$	

Though the dry deposition velocity varied with underlying ground surface conditions such as roughness, Stewart et al. (1983) also concerned the dry deposition velocity of sulfate (SO_4^{2-}) to every characteristics of the underlying ground surface, they suggested that a value of 0.005 m/s was taken for the lake or ocean, 0.003 m/sec for the metropolitan city, and 0.002 m/sec for cropland, woodland and grazing land. Waleck et al.(1986) estimated and parameterized sublayer and surface resistance for dry deposition of particulate sulfate from eddy correlation measurements which were carried out by Wesely et al. (1985). And they also showed that the dry deposition velocity for SO_2, SO_4^{2-}, and HNO_3 varied with difference of ground surface conditions. In this study, it was assumed that the dry deposition velocity for SO_2 and SO_4^{2-} varied with surface conditions as shown in Table 1. As the wet deposition, the below cloud scavenging was considered. The value of below cloud scavenging coefficient hve been suggested in several literatures until present (Eliassenn, 1982; Fisher, 1984; etc.), however it scatted widely from 10^{-5} to 10^{-3}/sec. In present study, it was assumed to $3x10^{-5}$/sec for SO_2, $1x10^{-4}$/sec for SO_4^{2-} for the precipitation over 0.1 mm/hour, as seen in Table 1. On the other hand, the transformation from SO_2 to SO_4^{2-} was considered, the rate is assumed to be 0.01/hour.

3. The Verification of the Transport Model

In order to verify the transport model, since the reliable data on emission inventory and observed wet deposition were provided at North America, the simulation was performed for winter season at North America. The emission inventory from National Precipitation Assessment Program (NAPAP) was used for the simulation. The calculation was carried out from December 4, 1984 to March 3, 1985. The calculated results of wet deposition of SO_4^{2-} were compared with observed wet deposition of SO_4^{2-} were compared with observed data from National Atmospheric Deposition Program and National Trend Net Work (NADP/NTN). Though dry deposition occurred probably when pollutants close encountered with sublayer just

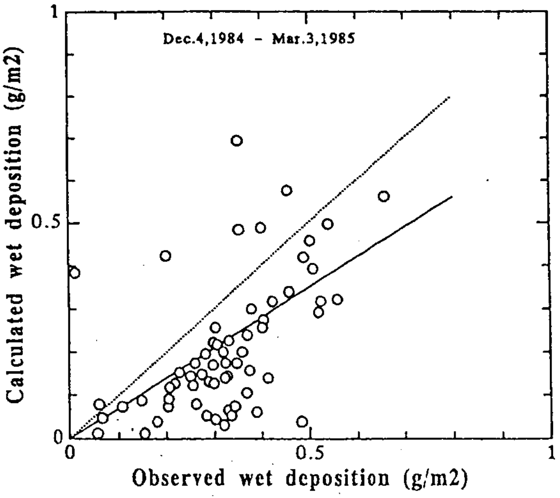

Fig. 2 Comparison between calculated and observed wet deposition.

above the ground surface, it depended on advection and diffusion for vertical direction. Consequently, the dry deposition occurred relatively close to emission. Meanwhile, in the case of wet deposition, pollutants in the atmosphere might be scavenged under some probability related with precipitation. Thus, the location of precipitating area were the most important for the distribution of wet deposition. The patterns of distribution between calculated and observed wet deposition for sulfate showed to be similar. Figure 2 shows scatter diagram between observed wet deposition of sulfate at each station and calculated one. The calculated wet deposition of sulfate was averaged in 31.8 km^2 (1/4 grid). The calculated wet deposition showed slightly under estimation. In this case the threshold of precipitation was set at 0.5mm/hour, hereafter it was reset at 0.1 mm/hour.

4. Simulation of the Transport Process in East Asia

The emissions for sulfur oxides in east Asia were distributed on the grids of the model by weighing on industrial activity and population. The simulation was performed for all the year aground 1985. The calculation was carried out by excepting the domestic emission of Japan for evaluating the contribution on acid deposition in Japan from the emission in vicinal countries. The calculated annual wet depositions of sulfur oxides at several areas in Japan were evaluated by comparing with observed wet deposition of sulfate. The calculated results are shown in Fig. 3, and observed wet deposition is shown in Fig. 4.

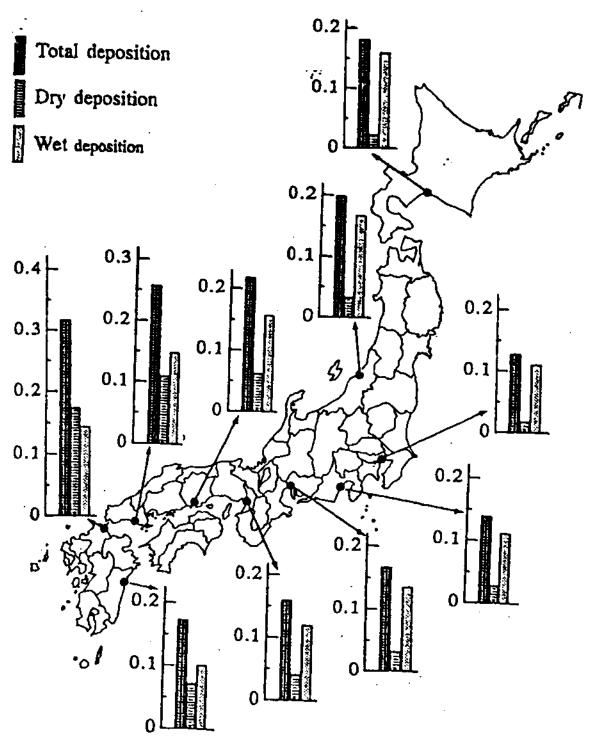

Fig. 3 Calculated wet deposition of sulfur oxides.

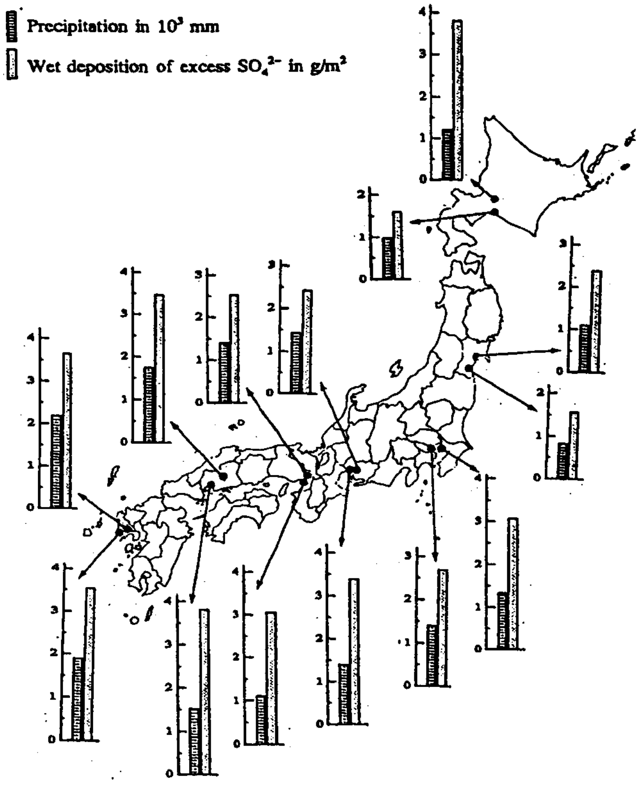

Fig. 4 Observed wet deposition of sulfate.

Since the present model used Lagrangian particle, it could be trace the path of particles. The contribution on acid deposition in Japan by the emissions in vicinal countries were investigated. Figure 5 shows the contribution on wet deposition at Kitakyushu area by the emission in vicinal countries for January and July.

5. Concluding Remarks

The numerical model of long-range transport have been developed, the numerical simulation of transport process on sulfur oxides was performed. The result of simulation showed that the wet deposition originated the emissions in vicinal countries was 0.1-0.3 g/m^2. This was about 1/10 lower than observed wet deposition on sulfate. The contribution on wet deposition originated the emission in foreign countries was recognized, It was from east part of China, Korea and Taiwan.

The present model did not include the **deposition** process of in cloud scavenging, it will be done in the future.

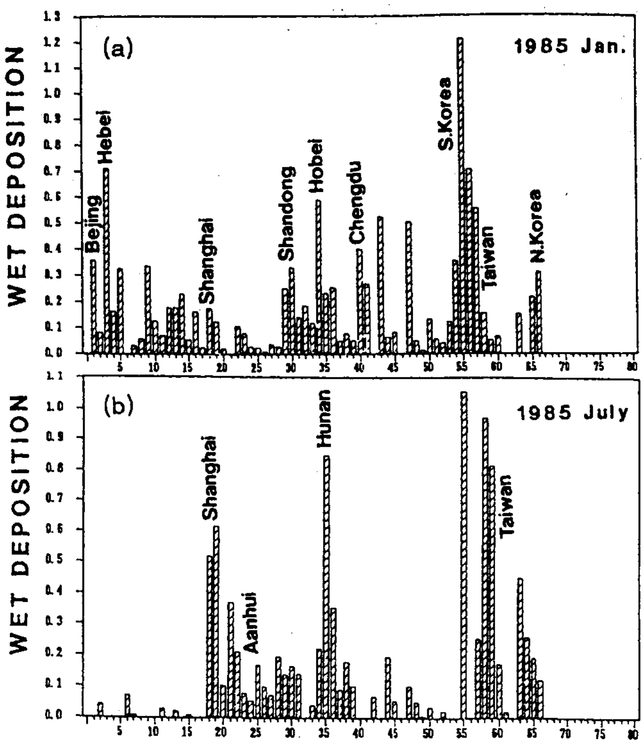

Fig. 5 The contribution on wet deposition from the foreign countries.

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