A NUMERICAL SIMULATION OF THE DISTRIBUTION OF ACID PRECIPITATION IN CHONGQING AREA OF CHINA

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ABSTRACT

A numerical model for the study of the regional acid precipitation is developed. The model consists of five parts: the distribution patterns of SO_2 concentration, the mesoscale flow fields, the parameterization of SO_2 transformation into $SO_4^{=}$, the parameterization of precipitation scavenging process, and the relationship between SO_2 content in precipitation and ground level concentration of SO_2 in the air. The distribution of SO_2, $SO_4^{=}$ and pH for all precipitations in Chongqing area during the period of July to October 1982 are simulated with the model. A comparison of the simulated results with experimental data shows that high SO_2 concentration centres correspond to low pH centres. The source of the acid rain in Chongqing area is local air pollution which is due to the lower effective stack height, low wind velocity in the area, basin topography, and the use of coal with high sulphur content. The mechanism for the formation of the acid precipitation here may be different from that in the United States of America and the Western Europe, where acid rain appears in the area far from pollution source.

I. INTRODUCTION

Over the past ten years, people have unceasingly paid great attention to environmental problems due to the medium-long range (100—1000 km) transport of pollutants in the air. Acid rain is one of the most striking environmental problems in the contemporary world, and has caused local disasters in some areas of China. It is one of the factors influencing the production of agriculture in the Southwest of China and the south of Yangtze River Valley. If we do not pay great attention to the problem, it will become a more serious problem.

Although the problem has now been given attention by some localities and departments in this country, and some precipitation acidity measurement and investigations of acid rain harmfulness are carried out, such problems as the formation cause of acid rain, the acid rain related to air pollutant, its effective range and sources, and so on, have not been tackled systematically.

Acid rain is a phenomenon of precipitation that involves microscopic and macroscopic physical and chemical processes of the pollutants in the air. Therefore it is an investigation topic with great variation both in temporal and spacial scales.

Based on the knowledge of transport, diffusion, removal and transformation of air pollutants, precipitation chemistry and cloud microphysics, and so on, a Lagrangian puff trajectory model is designed to get a quantitative and semi-quantitative knowledge about the sources, cause of formation, forecast of developing tendency for the acid rain and to develop strategies for the maintenance of the ecological equilibrium, rational depletion of energy sources, the best adoption of project outline of the environmental engineering, the protection of agricultural land resources, and so on.

II. ESTABLISHMENT OF THE MODEL

Generally speaking, a regional pollution model is bound to be of multiple sources. Moreover, because the pollutants undergo long-time and large-range travel in the atmosphere, the model must take account of the transport, diffusion, transformation, decay, wet and dry deposition (including precipitation physics and chemistry) between sources and receptor points, and the effect of complex terrain. The parameterization and modelling of the above physical and chemical processes will be discussed below.

1. *Concentration Distribution Patterns for the Pollutants with Complex Sources*

The key point of the model (Ren and Lei et al. 1981) is that the long-term average plume (or trajectory) from each pollution source is approximated by a series of Lagrangian puffs, the positions of which can vary with wind direction during the moving process, and hence the plume may bend. The concentration at a given point (*x*, *y*, *z*) is computed by the sum of all the contributions from the puffs to that point, i.e.,

$$C_q(x,y,z)=\sum_{i=1}^{M}\sum_{j=1}^{N}Q_{ijq}Ph(x-\bar{x}_j,y-\bar{y}_j)P_{zq}(z,t), \tag{1}$$

where subscripts $q=1$ or 2 represent SO_2 and $SO_4^=$, respectively, Q_{ijq} is the source emission rate of the qth species in the jth puff emitted from the ith source, M is the total number of the pollution sources, N is the total number of puffs for each plume, Ph and P_{zq} are the horizontal and vertical distribution functions of pollutant concentration, respectively. Ph is given by

$$Ph(x-\bar{x}_j,y-\bar{y}_j)=\frac{1}{2\pi\sigma_x\sigma_y}\exp\left\{-\frac{1}{2}\left[\left(\frac{x-\bar{x}_j}{\sigma_x}\right)^2+\left(\frac{y-\bar{y}_j}{\sigma_y}\right)^2\right]\right\}, \tag{2}$$

where $\bar{x}_j$ and $\bar{y}_j$ are the mean trajectory coordinates for the plume, σ_x and σ_y are the standard deviation of the puff trajectory. The vertical distribution functions of SO_2 and $SO_4^=$ P_{zq} are written as

$$P_{z_1}(z,t)=(4\pi k_z t)^{-1/2}\exp\left\{-\frac{(z-z_s)^2}{4k_z t}-\frac{2V_{d_1}t^{1/2}}{(\pi k_z)^{1/2}}-\Lambda_1 t-A_1 t\right\}, \tag{3}$$

$$P_{z_2}(z,t)=(4\pi k_z t)^{-1/2}\exp\left\{-\frac{z^2}{4k_z t}-\frac{2V_{d_2}t^{1/2}}{(\pi k_z)^{1/2}}-\Lambda_2 t-A_2 t\right\}, \tag{4}$$

k_z is the vertical eddy diffusion coefficient, which is regarded as a constant in the model and was given by Wisplaere (1980) as

$$k_z=ku_*z\cdot\exp\left[-7.8\left(\frac{zf}{ku_*}\right)^{0.764}\right], \tag{5}$$

k is the Von Karmen constant (0.36), f is the Coriolis parameter, u_* is the velocity scale in the surface layer (Lei and Den, 1986)

$$u_* = 0.335 + 0.31 z_0, \tag{6}$$

z_0 is the effective roughness length (Lei et al., 1981), Λ_1 and Λ_2 are the precipitation scavenging coefficient for SO_2 and $SO_4^=$ respectively, A_1 and A_2 are the decay constants for SO_2 and $SO_4^=$ respectively, V_{d1} and V_{d2} are the dry deposition velocity for SO_2 and $SO_4^=$, respectively, z_s is the plume effective emissive height. The height of the plume rise is computed by Briggs formula (Lei, 1983).

2. *Flow Field and Trajectory*

For finishing the concentration calculation mentioned above, the basic parameters σ_x, σ_y and $\bar{x}_j$, $\bar{y}_j$ must first be determined. The puff trajectory coordinates at each time are defined as

$$x_{jl} = u(x_{(j-1)l}, y_{(j-1)l})\Delta t, \tag{7}$$

$$y_{jl} = v(x_{(j-1)l}, y_{(j-1)l})\Delta t, \tag{8}$$

where Δt is the time interval between the two consecutive puffs, u and v are wind velocity components in the x and y directions, respectively, $j = 1, 2, \ldots, N$ are the end point of the trajectory at the time of $t = 1, \ldots, N\Delta t$ counting from the puff releases, and l is the label for each trajectory (or the plume formed by the puffs), the average coordinates of the trajectory can be described by

$$\bar{x}_j = \frac{1}{L}\sum_{l=1}^{L} x_{jl}, \tag{9}$$

$$\bar{y}_j = \frac{1}{L}\sum_{l=1}^{L} y_{jl}, \tag{10}$$

where L is the total number of trajectories. It is very clear that, for the computation of mean trajectory and σ_x, σ_y the key of the problem is that velocity components u and v are known at each point in the considered range. However, the density of network for operational wind measurement is far from enough. Therefore, it is important to obtain necessary data for each computation point from the observational data by objective analysis method. Besides, only those data of surface wind are available for most of the observation stations, so that it is important to obtain wind speed at different heights from the surface wind measurement, especially when the emissive source is at a higher level. A problem is how the computation formula of variation of wind speed with height is correctly selected. In the model, the empirical Ekman wind profils provided by Yuan and Lei (1982) is used

$$U_Z = 1.832[1 - \exp(-0.3218 z^{0.2695}) \cos(0.3218 z^{0.2695})] U_{10}, \tag{11}$$

where U_{10} is the wind speed at 10 m, the components of wind speed is

$$u_Z = U_Z \cos(270 - \theta), \tag{12}$$

$$v_Z = U_Z \sin(270 - \theta), \tag{13}$$

where $U_z = (u_z^2 + v_z^2)^{1/2}$, θ is the wind direction. The wind speed components at each mesh point are computed by objective interpolation formula (Ren and Lei et al., 1981)

$$u = \sum_{T=1}^{N1} \frac{u_{Z}l}{r_T^2} \Big/ \sum_{T=1}^{N1} \frac{1}{r_T^2}, \tag{14}$$

$$v=\sum_{T=1}^{N_1}\frac{v_z t}{r_T^2}\Big/\sum_{T=1}^{N_1}\frac{1}{r_T^2}, \tag{15}$$

where N_1 is the number of the objective interpolation point, r_T is the distance between observation point and mesh point.

3. *Parameterization of* SO_2 *Transformation into* $SO_4^=$

Because $SO_4^=$ in the atmosphere or raindrops mainly come from SO_2, for the computation of $SO_4^=$ concentration in the air or precipitation it is necessary to know Q_{ij_2} (e. g. the source strength). In the model $SO_4^=$ concentration is determined from the concentration of SO_2 at each time step and mesh point (being a function of time and space), which is quite different from the case of SO_2 where the concentration is determined by each emissive source. For Q_{ij_2} we adopt the method used in Europian regional model (Johnson, 1977)

$$Q_{ij_2}=\frac{3}{2}k_t\int_0^t c_1 V dt, \tag{16}$$

where k_t is the conversion coefficient of SO_2 to $SO_4^=$, V is puff volume at each time and is given by

$$V=\pi h R^2, \tag{17}$$

where R is a radius of the puff at any time t, h is the mixing layer depth. Supposing that the puff diffusion follows the Fich rule in the process of transportation, then R can be given approximatively as

$$R=(R_0^2+k_0 t)^{1/2}, \tag{18}$$

where k_0 is the horizontal eddy diffusion coefficient, R_0 is the initial radius of the puff. Substituting (18) and (17) into (16), we get

$$Q_{ij_2}=\frac{3}{2}\pi h k_T\left[R_0^2\int_0^t c_1 dt+k_0\int_0^t c_1 t dt\right]. \tag{19}$$

After numerically integrating (19), the source emission rate Q_{ij_2} can be obtained at each puff passing point.

4. *Parameterization of Precipitation Scavenging Process*

The determination of precipitation scavenging coefficients Λ_1 and Λ_2 is one of the key problems in the acid rain model. In this paper, the method computing scavenging coefficient due to Scott (1982) is used. Scott indicated that his results could be used to mesoscale or regional scale problems. Considering a series of microphysical process of precipitation, he got the scavenging coefficient Λ

$$\Lambda=1.26J(z)^{0.78},\ (\mathrm{h}^{-1}) \tag{20}$$

where 1.26 is a constant with dimension, $J(z)$ is a precipitation rate (mm/h), which is a function of height to be described as

$$J(z)^{0.22}=J(g)^{0.22}-3.1\cdot10^{-4}\bar{m}(z-z_g), \tag{21}$$

where $\bar{m}$ is the average cloud water concentration (g/m^3) between the ground and height Z, $J(g)$ is the precipitation rate on the ground (mm/h), Z_g is the elevation of the surface (m). Substituting (21) into (20), we obtain

$$\Lambda=1.26[J(g)^{0.22}-3.1\cdot10^{-4}\bar{m}(z-z_g)]^{39/11}. \tag{22}$$

5. *Equilibrium Relationship between* SO_2 *Content in Precipitation and Ground Level Concentration of* SO_2 *in the Air*

A trial is made to relate the pollutant concentration in the air with the pH value in the precipitation. In the SMICK model of Drewes (1982), the chemical kinetics incorporating with transport in the atmosphere are analysed, and two control equations for the pollutant behavior in the aqueous phase and the gaseous phase are explicitly presented. Good results have been obtained in practical applications, and it has been indicated that the aqueous phase concentrations are influenced by the gaseous values.

Recently, Pena (1982) has also investigated the relationship between the SO_2 concentration in rain samples and the surface SO_2 concentration in the air, as well as the relationship between the SO_2 concentration and pH value. For the relationship between the SO_2 concentration in the atmosphere and aqueous phases in the precipitation process, the following formula is given by Hales (1979)

$$c_x=\frac{c_y}{H}+\{-[H_3O^+]_{ex}+\sqrt{[H_3O^+]^2_{ex}+4k_1c_y/H}\}/2, \tag{23}$$

where $[H_3O^+]_{ex}$ (mol l^{-1}) represents the concentration of hydrogen ion donated to the precipitation. As a first approximation, it is related to pH value by

$$[H_3O^+]_{ex}=10^{-PH}. \tag{24}$$

Substituting (24) into (23) and using expression $c_x=H'c_y$, (23) becomes

$$c_yH'=c_y/H+\{-10^{-PH}+(10^{-2PH}+4k_1c_y/H)^{1/2}\}/2, \tag{25}$$

where c_y is SO_2 concentration in the air (mol L^{-1}), H' is Henry's law constant for SO_2, which, as given by Fisher (1982), is dependent on the acidity of raindrops:

$$H'=H_0(1+k_1/10^{-PH}), \tag{26}$$

Where H_0 is a constant to be determined, k_1 is a parameter (mol L^{-1}) depending on temperature. It can be expressed as

$$k_1=\exp(2.34-0.0224T). \tag{27}$$

Eq. (27) is obtained by a regression analysis with the data given by Hales (1979), with a correlation coefficient $r=0.9999$; T is atmospheric absolute temperature, H is a parameter (dimensionless) depending on temperature and is given by

$$H=1.015\cdot10^{-25}T^{9.499}. \tag{28}$$

It is also obtained by a regression analysis with the data given by Hales (1979), with a correlation coefficient $r=0.998$. Based on Eqs. (25) and (26), an empirical formula for the relationship between the pH value in precipitation, SO_2 concentration in the atmosphere, and atmospheric temperature is obtained by using both gradual approach and nonlinear regression analysis, i.e.

$$pH=(40.606-6.464\ln T)c_y^{-0.04817}. \tag{29}$$

The unit of c_y in Eq. (29) is mg/m^3, different from that in Eq. (25). From Eq. (29) it can be seen that at given atmospheric temperature and other parameters, substituting concentration results computed by (1) into Eq. (29), the corresponding space distribution of pH is obtained. Thus it is very convenient to connect pH in the rain water with pollutant sources and with physical and chemical processes in the air.

III. SELECTION OF PARAMETERS AND SOURCE OF DATA

The mathematical model obtained in the above paragraph can be applied to general cases. If the model is used for a specific area, the characters of each area must be considered, i.e. the data and parameters in the model may be different. In this section, we shall give examples of the acid rain in Chongqing area, the input data and parameters in the model are explained as follows.

1. *Case Selection*

Six continuous precipitation processes (1, 16—18 July, 21—24 August, 7—12, 15—19 September and 3—5 October, 1982) were selected as calculating examples (about 408 hours) in a range with an area of 75 × 150 km^2 (including all the suburban counties of Chongqing area and 10 pH monitoring stations). Puffs are emitted hourly, and the total 408 puffs are emitted for each pollution source. The computing mesh interval is $\Delta x = \Delta y = 3$ km.

2. *Pollution Sources*

In the computing process, pollution sources are SO_2 from power plants, industries and residential areas in six areas of the city of Chongqing. The plume effective emissive height for all the industrial sources (except power plants) is taken as 50 m. The residential source of each area is converted to four point sources with a height of 10 m. Thirty-three point sources are computed in the model.

3. *Meteorological Parameters*

The data of wind speed, wind direction, temperature and precipitation were obtained from 8 surface meteorological observation stations. The meteorological parameters are put into the model at each hour, so that with the model calculation one can track the trajectory of the puff once an hour.

The height of the mixing layer h was obtained from radiosonde data provided by Chongqing Meteorological Observatory, with an average height of $h = 1150$ m over the period of model calculation.

4. *Other Input Parameters*

In addition to the above-mentioned data and the parameters given by Ren and Lei et al. (1981), the following parameters are also used in the calculation:

In order to take into account the large terrain ups and downs and dense buildings in Chongqing City the effective roughness length z_0 is selected as 2 m, which had been obtained by Chongqing Institute of Environmental Science and Monitoring.

The Coriolis parameter in the middle-latitude area is $f=10^{-4}\ s^{-1}$. As for the selection of the conversion ratio of SO_2 to $SO_4^=$ based on the data provided by Durran (1979), $k_t=1$—13% h^{-1} for city or humid environment; therefore, we take $k_t=0.1\ h^{-1}$ for Chongqing, an area humid and rough. Λ_1 is given by Eq. (22), $\Lambda_2=0.1\Lambda_1$, $m=0.3\ g/m^3$ (for warm cloud precipitation). According to the observed average value of pH over Chongqing and empirical formula $H' \approx 1.2\cdot 10^{pH}$ given by Fisher (1982), H_0 in H' is taken as 26.

IV. RESULTS

Using the model and the data introduced in previous sections, the space distribution of SO_2, $SO_4^=$ concentrations, and the pH values, as well as the trajectory of acid rain have been numerically computed for 6 representative continuous precipitation processes. The results are given in Figs. 1—4, respectively.

Fig. 1 shows the trajectories of acid rain in Chongqing area. The region enveloped by the dotted line in the figure is the source region (namely, Chongqing City area). We only draw 5 representative average trajectories for different time periods (dividing the data of 408 hours into 4 periods for calculation). The characters that can be seen from the figure are: (1) some of the pathes, such as the average trajectory of the 4th source in the 3rd period 3(4), turn around, pollutant emitted from the source will return back to the source point after 26 hours; (2) non of the 4 pathes is in a straight line, with eviden wind direction shear; (3) the mean trajectory directions for the four periods differ greatly from SW to NE in period one, from NW to SE in period two, from NE to SW in period four, and more complex in period three. These characters indicate that the acid rain in Chongqing City has some local features, with an overall tendency from N to S, a phenomenon which can be seen from wind direction frequency in the period, with 50.24% of the wind direction in the range of 315—45° and 31.1% in the range of 157.5—225°, and the rest is near east and west directions.

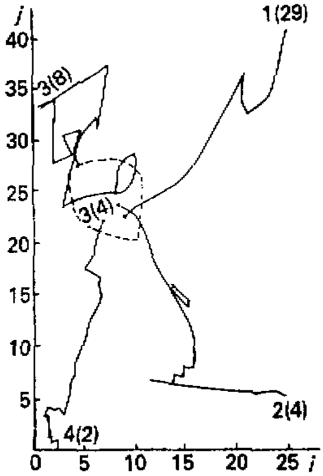

Fig. 1. Trajecfories of acid rain in Chongqing area.

The average concentration of SO_2 in the whole calculating period is shown in Fig. 2. There are three centres with the concentrations exceeding 0.1 mg/m³. The enveloped region of the three centers coincides with the Chongqing City area. The equi-scalar lines of concentration are ellipses with the long axis running from NNE to SSW. These results agree with wind direction frequency and the calculated trajectories. In order to test the reliability of calculated results, we have compared the computed values with the measured mean concentration of SO_2 in summer in Chongqing. The measured mean value is 0.17

mg/m^3, while the calculated one is 0.14±0.035 mg/m^3, a bit lower than the former. There are possibly two reasons: one is that the measured value is made in a few fixed points, while the calculated value is the average of all points with 3 km intervals, so that the calculated space average is more representative; the other reason is that the measured value is obtained under all meteorological conditions, while the computing condition is for precipitation process. Because of precipitation scavenging, the SO_2 concentration in the atmosphere is lower than that in the case without precipitation. Therefore, the calculated value should be lower than the mean measured value.

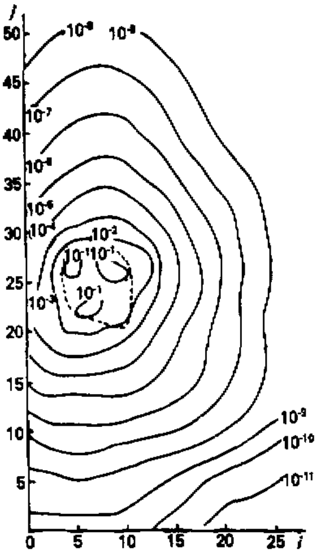

Fig. 2. Distribution of SO_2 concentration (mg/m^3) in Chongqing area.

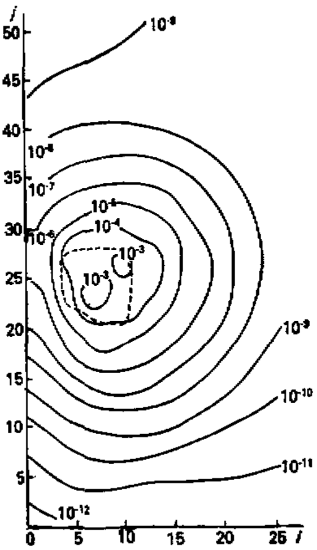

Fig. 3. Distribution of $SO_4^=$ concentration (mg/m^3) in Chongqing area.

Fig. 3 shows the mean concentration of $SO_4^=$. The basic distribution pattern and alignment are the same as SO_2, but the concentration of $SO_4^=$ is less than SO_2. The difference between SO_2 and $SO_4^=$ concentration depends on the direction and distance. The results coincide with those by Ren and Lei et al. (1981). The ratio of SO_2 concentration to $SO_4^=$ concentration is two orders of magnitude near the source, 1.38 at the most NE direction in the model, 1.65 at the most SE direction, 22.4 at the most NW direction, and three orders of magnitude at the most SW direction, depending on the wind speed in each direction.

Fig. 4 shows the distribution of average pH in rain. Supposing 5.6 is the standard pH value for acid and non-acid rain, Fig. 4 indicates that the range with acid rain due to the pollution sources in Chongqing City itself, is a circular area with the center at the central city, with a radius of 25 km, larger than the city's radius by 15 km, and the center of acid rain overlapps with the high SO_2 concentration center.

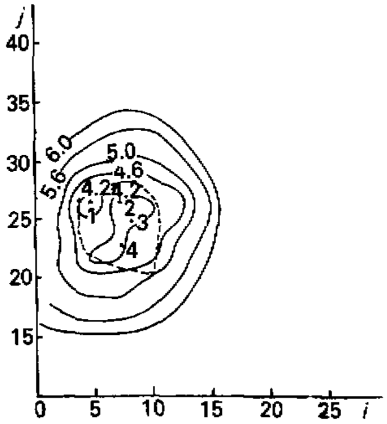

Fig. 4. Distribution of pH in rain in Chongqing area.

For the whole calculating range and period, we applied the rainfall amount weighted method to get the overall average pH value for each station. There are nine observation stations for pH, only four of them are located in Chongqing City. Only the effect of Chongqing City area pollution sources is considered in this study, so that we only compare the calculated value with the data of the four city stations. One of them is Sha Pingba station, where the mean pH = 3.83, while the lowest calculated pH is about 4. The second station is the Institute of City Monitoring, where the mean pH is 3.96, while the calculated value is near 4.1. The third station is Yangtze Power Station, where the mean pH is 3.99, while the computed value is about 4.2. The last station is Jiu Longpo, where there are only two observational data for pH, one of them is 4.68, but the calculated value is about 4. From the above results it can be seen that the lowest value of pH due to the pollution sources in the city is about 4.0, slightly higher than the observational value. This is probably due to the pollution sources outside the city. On the whole, the modeling results are in good agreement with observations.

In summary, the acid rain over the Chongqing area is caused mainly by its own pollution sources. One of the reasons is the lower emissive height of pollution sources, and the other is the weak wind and the unstable wind direction with the pollutants traveling at an average speed of only 1.26 km per hour. Therefore, the mechanism for the formation of acid rain there may be different from that in the United States of America and the Western Europe, where acid precipitation appears in those areas far from pollution sources.

V. SUMMARY

From the above analysis we can see that, as a study of acid rain problem, the model has the following characters:

1. It can provide information on the space distribution of SO_2, $SO_4^=$ concentrations, and the relative importance of diffusion, transport, dry and wet deposition, removal, transformation, and decay in the formation of acid rain, for calculating the trajectory of acid rain.

2. It relates the pollution source strength with the pH value in precipitation, which is

helpful for the study of the sources of acid rain. The results can provide a scientific basis for the control of acid rain.

3. Processes of atmospheric chemistry and atmospheric physics, macroscopic and microscopic processes are incorporated into the model, although some of them are only considered roughly. Mathematical modeling has a good prospect as a tool in studying acid rain, which is a comprehensive topic.

4. With the knowledge of the present energy depletions and future development, the model can be used to predict the tendency of the acid rain.

In short, mathematical modeling is a useful tool for the study of acid rain. Even with the help of rather rough data, it is still useful for solving certain problems, especially for strategic considerations. With further development in studies of regional acid rain, especially precipitation chemistry and physics, this model will play its potential role.

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