

## RECONSTRUCTION AND NUMERICAL ANALYSIS OF THE POLLUTANT CONCENTRATION FIELDS IN GEOGRAPHICAL REGIONS

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*We present here the Eulerian model of the vertically averaged sulfur dioxide stationary transfer over Europe. This model is based on numerical solution of the transfer equation of (balance) atmospheric pollutants. We have obtained the "climatic" stationary concentrations of sulfur dioxide in air over Europe for different seasons assuming the process to be stationary. In so doing, we used the climatic data and considered the advective transfer processes, the vortex turbulent diffusion, and dry and humid reconstruction. The chemistry is presented in a simplified form. To create the numerical algorithm we used the finite-difference approximation.*

The environmental pollution and particularly the air pollution is among most important environmental problems. Most dangerous air pollutant is sulfur and its compounds, principally, sulfur dioxide (SO<sub>2</sub>). The impact of this gas on the air quality has been discussed as long ago as in the seventeenth century. About one hundred years ago sulfurous gas was mentioned as a possible reason of wood's degradation in Germany. For certain reasons the attention to this gas does not weaken until now. First, sulfur dioxide, as well as nitrogen oxides, cause the acid rains, which remain a regional problem, Ref. 7. Second, the investigators of clouds offer a hypothesis that in the regions far away from industrial zones the condensation nuclei mostly consist of sulfurous components. In one way or other, it is not known as yet, but it is hardly possible to answer this question without studying sulfur compounds in the atmosphere. Third, the question on the climate connection with sulfate aerosol remains a disputable one. One supposes, that the direct influence of the sulfate aerosol is in the solar radiation reflection, and indirect influence in the modification of both the albedo of clouds, and their amount. Fourth, some authors pointed out, that the oxidation of sulfur dioxide dissolved in droplets of clouds, is the essential sink for photo-oxidants particularly of the ozone. Taking into account a huge emission of SO<sub>2</sub>, it is the point to consider this process at calculations of the regional ozone amount.

One of the most effective methods enabling the consideration of the above mentioned problems, is the numerical simulation. The simulation of air pollution transfer is being done for some tens of years already. Nowadays two basic types of models (Lagrangian and Eulerian models) are normally used. In the former models there have been carried out calculations of the diffusion, transformation and reconstruction of

pollutant along the motion trajectory of a unit air volume. The trajectory is found on the basis of observed or forecasted wind field. For Eulerian models the solution of the problem of transfer and transformation of a pollutant is performed on the stable geographical grid. Thus the region considered is divided into two- or three-dimensional cells of the grid, and the solution of the equation or the system of the equations is performed for each cell.

### DESCRIPTION OF THE MODEL FOR CALCULATION OF THE SO<sub>2</sub> CONCENTRATION

In this paper we offer the model of sulfur dioxide transfer over Europe. Practically all models, that have been developed up to now, are intended for calculation of transfer for any determined time interval. In this paper we suggest to obtain the concentration fields using the climatic data. We suppose that the process of the pollutant emission and its consequent transfer and transformation is stationary.

This model is based on the equation of transfer (balance) of atmospheric pollutants. In the general view it can be written as<sup>4</sup>

$$\frac{ds}{dt} + w_a \frac{\partial s}{\partial z} = k_1 \Delta s + \frac{\partial}{\partial z} k_z \frac{\partial s}{\partial z} + Q + R - W. \quad (1)$$

Here  $s$  is the volume concentration of the pollutant,  $w_a$  is the vertical velocity of the pollutant itself,  $k_1$  and  $k_z$  are the turbulent coefficients at moving along the horizontal and vertical directions, respectively,  $Q$  characterizes the inflow of the pollutant,  $R$  is the modification of the pollutant amount at the expense of chemical transformation,  $W$  describes the processes of the pollutant removal.

By the processes of the pollutant removal we understand first of all the dry removal and the humid

washout. The former process is nothing but the adsorption of molecules of the pollutant particles on the surface, for instant, by soil, water or vegetation. The theoretical aspects of this process are presented in a series of papers, for example, in Refs. 5, 7, 8, 12, and 14. The process of the humid washout is that the pollutant is dissolved in droplets of atmospheric water, and the content of the air pollutant is thus reduced. It can be represented in two ways (the in-cloud and the under-cloud washout). In the first case it is meant, that the substance washed out either participates in formation of the cloud droplets, or is captured by the cloud droplets, while they do not drop as rain. In the second case the substance is captured by an incident droplet of the rain. The mathematical description of the removal processes is given in a series of papers, for example, in Refs. 8, 12, and 13.

As has already been mentioned above, the problem was considered in the assumption that stationarity conditions already exist. Thus, we do not take into account local derivatives with respect to time.

To solve this problem we have proposed to use the technique developed and described in Refs. 4 and 5. According to these papers, the evaluation of the regional pollution level is carried out for the region within the mixing layer, inside which the authors calculated the average concentration of a pollutant. To do this, Eq. (1) was vertically averaged, i.e., Eq. (1) was integrated over height from 0 up to  $H$ , where  $H$  is the height of the mixing layer, and we introduced the average values of the concentration, velocities and sources (sinks)

$$\begin{aligned} \bar{s} &= \frac{1}{H} \int_0^H s(z) dz, & \bar{u} &= \frac{1}{H} \int_0^H u(z) dz, \\ \bar{v} &= \frac{1}{H} \int_0^H v(z) dz. \end{aligned} \tag{2}$$

At averaging we used the following conditions on the upper and lower boundaries:

$$\begin{aligned} k_z \frac{\partial s}{\partial z} &= 0, & w_a &= 0, & w &= w_H & \text{at } z = H; \\ k_z \frac{\partial s}{\partial z} - w_a s &= \beta s - f_0, & w &= 0 & \text{at } z = 0, \end{aligned} \tag{3}$$

where  $f_0$  is the rate of the pollutant emission from the surface source,  $\beta$  is the rate of the pollutant absorption by the Earth's surface.

At averaging, apart from the averaged concentration, there occur the values  $s_0$  (the near surface concentration) and  $s_H$  (the concentration on the upper boundary of the mixing layer). Those can be expressed as follows:

$$s_0 = \bar{s} \alpha, \quad s_H = \alpha_H \bar{s},$$

$$\text{where } \alpha = \frac{s_0}{\bar{s}}, \quad \alpha_H = \frac{s_H}{\bar{s}}. \tag{4}$$

The values  $\alpha_H$  and  $\alpha$  are obtained from semiempirical relations, Ref. 5. When realizing this scheme we considered that the problem is stationary and also that  $k_x = k_y = k$ .

Keeping that in mind, and taking into account Eqs. (3) and (4), we obtain the final equation used in the model

$$u \frac{\partial s}{\partial x} + v \frac{\partial s}{\partial y} - k \Delta s + \sigma S = F. \tag{5}$$

In Eq. (5) the sign of the averaging is omitted,  $F$  designates the sources,  $\sigma$  is the parameter which takes into account the processes of pollutant removal from atmosphere, i.e., the dry absorption, the influxes through the upper boundary, the humid reconstruction, the chemical reactions, i.e., we can write (see Ref. 4)

$$\sigma = \sigma_1 + \sigma_2 + \sigma_3 + \sigma_4. \tag{6}$$

Here  $\sigma_1 = \alpha_0 \beta/H$  is the parameter, determining the dry sedimentation,  $\beta$  is the rate of dry sedimentation,  $\sigma_4 = \alpha_H w_a/H$  characterizes the influx of the substance that has its own velocity towards the surface. But in our case the considered substance is a gas, therefore, its own velocity may be ignored, therefore  $\sigma_4$  will not be mentioned further. The parameter  $\sigma_2$  characterizes the humid washout of a pollutant, and  $\sigma_3$  determines the chemical transformations.

### MATHEMATICAL REALIZATION

We solved the above stated problem by the finite-difference method. The specific feature of the elliptic equation (5) is the presence of the first order derivatives.

According to Ref. 10, it is undesirable to approximate the derivatives by central differences, otherwise it is necessary to consider the coefficient of the derivatives of the second order (in our case) to be too large. Therefore, we applied the directional differences. According to Refs. 6 and 10, we used the following approximations of the derivatives:

$$\begin{aligned} u \frac{\partial s}{\partial x} &= u^+ s_x + u^- s_{\bar{x}}, \\ \text{where } u^+ &= \frac{u + |u|}{2}, \quad u^- = \frac{u - |u|}{2}, \\ s_{\bar{x}} &= \frac{s_{i,j+1} - s_{i,j}}{h}, \quad \text{and } s_x = \frac{s_{i,j} - s_{i,j-1}}{h}. \end{aligned} \tag{7}$$

Similarly there was represented the approximation for the term with the rate  $v$ . Laplacians were written in a rather usual way

$$\Delta s = - \frac{4s_{i,j} - s_{i,j+1} - s_{i,j-1} - s_{i-1,j} - s_{i+1,j}}{h^2}. \tag{8}$$

Here  $h$  is the grid step.

However it is necessary to keep in mind the fact that when using the directional finite differences there occurs the computing viscosity resulting in smoothing out of fields, Ref. 6.

In this model the boundary conditions were given as follows:

$$S_b = \text{const} , \quad (9)$$

where by the symbolic "const" is implied the concentration values obtained from observations or from any other model. At numerical calculations we took this value to be equal to zero in the our model.

The problem was solved by the iteration method.

### INITIAL PARAMETERS AND THE AREA USED IN CALCULATIONS

The  $\text{SO}_2$  concentration was calculated over the territory of the West Europe and European part of the former USSR on the model grid of Gidrological Meteorological Center,<sup>3</sup> with the step of 150 km. The whole calculation area was covered by a grid of  $35 \times 37$  cells. For each cell we calculated the average annual emissions of sulfur dioxide. The emission power in each cell was calculated by the linear interpolation method using the data of the EMEP joint program (Cooperative Monitoring and Evaluation Program on Transmission of Air Pollutants in Europe) for observation and evaluation of air pollutant propagation at large distances over Europe.

The average wind in the layer is calculated according to Ref. 5 by the following formulas:

$$\bar{u} = c_1 \bar{u}_g + c_2 \bar{v}_g , \quad \bar{v} = c_1 \bar{v}_g + c_2 \bar{u}_g . \quad (10)$$

Here  $c_1 = 0.914$ ,  $c_2 = 0.096$  are the layer' mean values of the geostrophic wind  $u_g$  and  $v_g$  were calculated from the geopotential fields on the isobaric surfaces of 1000 and 850 mB by the polynomial interpolation method. We used the data on the geopotential from Ref. 1. As we used the monthly mean values of geopotentials, the values of the wind also were monthly average.

The mean values of the mixing layer heights were set according to climatic evaluations presented in Ref. 9, and were taken identical in all cells of the grid, though actually the values  $H$  should vary spatially.

The parameter  $k$  is the coefficient of the vortex turbulence (the average scale turbulence). By the vortex turbulence we imply the interchanging of significant air volumes as a result of the vortex motion along the vertical axes, the horizontal sizes of vortices do not exceed  $4Dh$  ( $Dh$  is the grid step). According to Ref. 3 this coefficient may be parametrized in the method proposed by V.A. Schneidman

$$k = gL_{\min}^2 |\Omega| , \quad (11)$$

where  $|\Omega|$  is the modulus of the vortex rate,  $L_{\min}$  is the minimum wavelength, which is described explicitly by

the model ( $L_{\min} = 4Dh$ ),  $g$  is the empirical coefficient ( $g = 0.18$ ). In reality, the vortex turbulence coefficient is set at calculations, as a rule. When solving this problem we produced its evaluation, then it was set initially to be identical for all cells of the calculation area.

At the problem realization the dry sedimentation and the humid washout parameters were identical on the whole calculation area.

The chemical transformation of pollutant was set in a rather simple way. It was suggested, that the pollutant portion which has been emitted to the atmosphere, immediately transforms into a sulfate. According to Ref. 11 this portion is 5–15% of the initially emitted into the air.

The parameters used in calculations, are shown below.

The turbulence coefficient, $K$	$10^5 \text{ m}^2/\text{s}$
Height of the mixing layer, $H$	1000 m (January)
	1500 m (July)
	1250 m (April, October)
Parameter $\alpha_0$	1.72
Rate of dry sedimentation, $\beta$	1.0 cm/s
Dry sedimentation coefficient, $\sigma_1$	$1.72 \cdot 10^{-6} \text{ 1/s}$
Washout coefficient, $\sigma_2$	$1.0 \cdot 10^{-5} \text{ 1/s}$
Portion of pollutant, transformed into a sulfate	10%

### DISCUSSION OF THE RESULTS OBTAINED. FURTHER IMPROVEMENTS OF THE MODEL NEEDED

At achieving the above stated task, the numerical algorithm for solution of the equation of pollutant transfer was developed. Based on climatic data, we have calculated the concentration fields of sulfur dioxide. Nevertheless, much more careful preparation of the initial data, particularly dry and humid reconstruction parameters, meteorological information, and chemical transformations are required.

As a result, it is possible to present the calculated fields of sulfur dioxide concentration above Europe for different months of a year. Those are shown in Figure 1.

Figure 1 shows at first, that the obtained spatial inhomogeneities of sulfur dioxide concentration reflect the distribution of its sources. The maximum values coincide with the geography of large industrial centers and regions, and also of cities with large population. The regions of the central part of Germany, Donbass, London, and Moscow are rather precisely exhibited. Second, we see the manifestation of the western transfer influence.

It is also seen, that the concentration values are largely similar for all four months. Most likely, this result is connected with the fact that the spatiotemporal variability of parameter's set, i.e., the precipitation and the related coefficient of humid removal, the dry precipitation, the mixing layer height, were not taken into account.

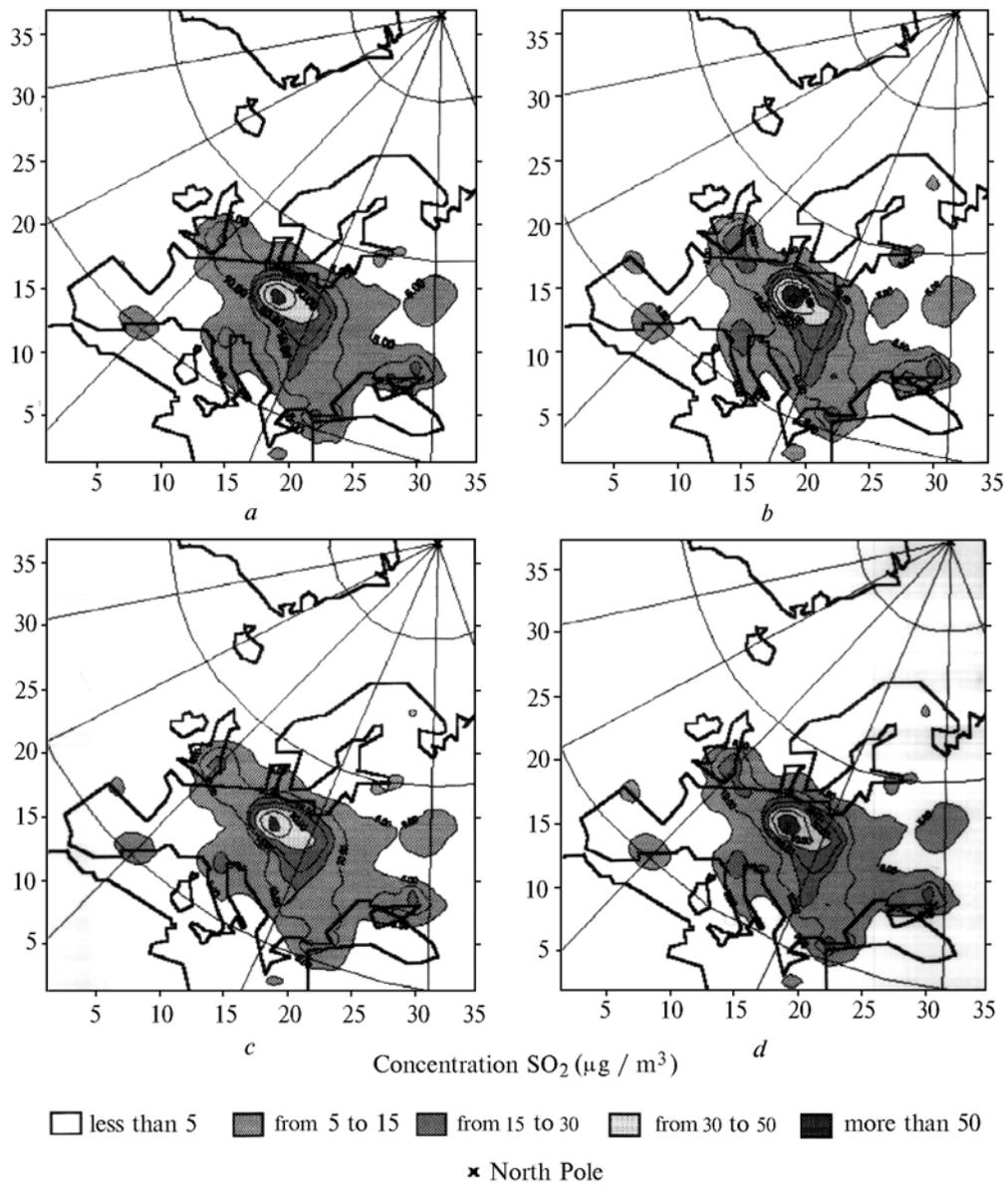


FIG. 1. Concentration of sulfur dioxide over Europe in different months: January (a), July (b), October (c), and April (d).

Thus, this model requires certain improvements. Among the basic ways for the improvements, it is possible to mention the following ones.

First, to consider the spatial inhomogeneities and seasonal variability of the precipitations.

Second, to consider the spatial modifications of the dry reconstruction coefficient, as it first of all depends on the type of the underlying surface.

Third, to consider a variability of the mixing layer height.

Fourth, to think about a more detailed description of the chemical transformation processes.

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