The empirical model of the interaction of aerosol with the chemical impurities under urban conditions

O.G. Khutorova, G.M. Teptin, and A.F. Latypov

Kazan State University

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We present an empirical model of interaction among different contaminating impurities in the atmosphere. This problem was solved based on the correlation analysis of long-term series of data on the concentration of aerosol, carbon monoxide, nitric oxide, nitrogen peroxide, and so on, including meteorological quantities. Three groups of impurities have been revealed depending on their interrelation character. It has been found that besides meteorological conditions the atmospheric turbulence plays also important role in the interaction of chemical impurities with the aerosol.

Since 1995 the measurements have been being carried out in Tatarstan of the contaminating species content in air under the program of regional environmental monitoring. At present, the observation points are located in Zelenodolsk (54°N, 49°E) and Almet'evsk (53°N, 51°E).[1]

The measurement complex comprises the instruments manufactured in Germany. The species that are monitored at these observation points are nitric oxide, nitrogen peroxide, sulfur dioxide, carbon monoxide, and aerosol.[2]

All the quantities were measured at 2.4 m height with the time resolution of 1 minute. More than 600 thousand measurements of every species were conducted per week. The data compiled on every species made up long-time series. As an example, we use in this paper the data bank on all the above-mentioned quantities measured every minute over the period from the 1st until 14th October 1997 at the station in Almet'evsk (53°N, 51°E).

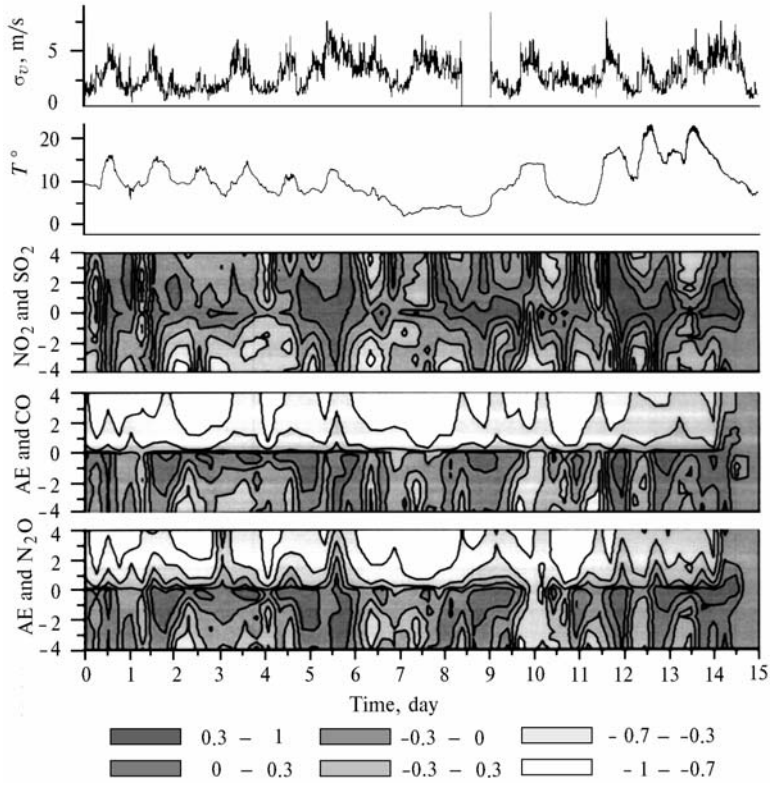

Fig. 1. Time series of the turbulent variation intensity, the wind velocity σ_v, the temperature T, the cross section of cross-correlation functions of aerosol (AE) and gaseous species on the plane (time, in days along the x-axis; time delay, hours – y-axis).

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In this paper, the problem is formulated of developing a statistically reliable empirical model of interactions among different contaminating species including aerosol. This problem was solved using cross-correlation analysis of long-time series of data on aerosol concentration, carbon monoxide, nitric oxide, nitrogen peroxide, and soot, including meteorological quantities. The method of the cross-correlation analysis assumes determination of the correlation coefficient between two steady-state time series $x(t)$ and $y(t + L)$ as a function of time lag L between the series.[3] Some characteristic time scales can be isolated in the energy spectrum of variations of the meteorological quantities.[4] These scales with the duration shorter than 10 minutes correspond to the acoustic short-period internal gravitational waves and turbulence. To evaluate the intensity of turbulent processes, we calculated the rms deviation of the wind speed within 10-min wide sliding time window because it is the turbulence that produces the main contribution to the wind speed variance.

Since the processes determining the interaction of aerosol and gaseous species have too complicated time variations, we analyzed the dynamic correlation functions constructed for a sliding time window. The time window of 12 hours was shifted along a long series of every minute measurement with the step of 6 hours. Inside the time window, the cross-correlation functions were calculated between the two time series analyzed with a maximum delay of 240 min. The results are plotted in the form of contour cross sections in Fig. 1. Figure 1 also shows the plots of temperature and intensity of turbulent variations, estimated from the variance of wind speed.

Three groups of parameters were determined based on the type of correlation. The processes determining the atmospheric emissions and the transfer of CO, NO_2, and NO have a strong interconnection (the coefficient of cross-correlation is 0.7–0.9). The cross-correlation functions of the concentration of these species are symmetric and decrease with the increasing delay of time series relative to each other (Fig. 2).

Aerosol has a significant correlation with gaseous components (0.5–0.8). In this case, the cross-correlation function is significant at a negative delay of measurement time series, i.e., at aerosol concentration advance. This is probably caused by the following circumstances. First, the rate of aerosol sedimentation in the ground layer is higher than that for CO and NO; secondly, the time of transition processes of gas emission from aerosol particles with the increase of insolation may also be important. Maximum values of cross-correlation are observed at advancing the aerosol time series to about 20 min. This fact indicates that the most probable time of transition processes of transformation of inhomogeneities of the concentration of suspended particles is 20 min. Figure 1 shows daily variations of cross-correlation functions. From this figure, we notice that during daytime even at two hour interval the correlation between time series of aerosol and gases is significant (0.3–0.1). At the same time, the maximum temperature (minimum humidity) and maximum intensity of turbulence are observed. The process, determining the interconnection of aerosol and gases, is evidently related to the atmospheric dynamics since at minimum temperature values but extended in time increase of turbulent intensity (5, 6, 9, 13, 14 days) the interconnection increases between the aerosol concentration and the concentration of CO, NO_2, NO, SO_2.

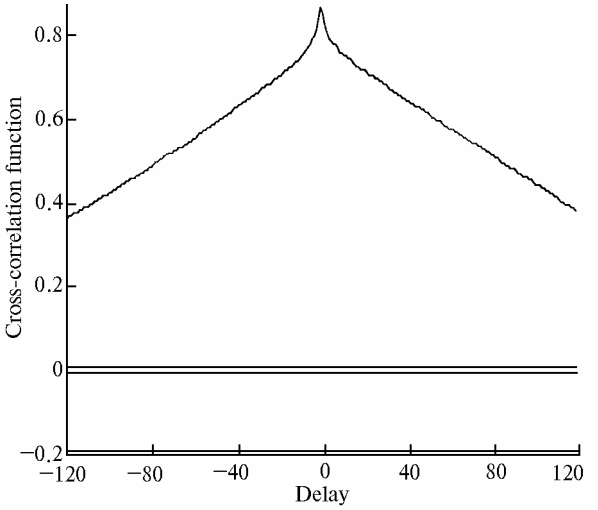

Fig. 2. Cross-correlation function of CO and NO concentration.

To the third group of the interaction type we attribute the connections of SO_2 concentration with the CO, NO_2, and NO concentration. At strong correlation (0.6–0.8) the values of cross-correlation function are significant at positive delay of time series of measurements, i.e., when advancing the SO_2 concentration (Fig. 1).

Different models of the admixture transfer show that the turbulence, planetary and mesoscale waves are of a considerable importance.[5–7]

The observations of impurity concentration at several spaced stations have made it possible to find the rates of the impurities transfer in the city; for suspended particles and CO the rate is, on the average, 0.4 m/s, and for NO and NO_2 – 0.5 m/s, while the mean value of wind speed for the observation period is 3.7 m/s.

In conclusion, it may be stated that, besides the meteorological conditions, the turbulent and mesoscale wave processes are of crucial importance in formation of the interconnections of suspended particles with gases because they can contribute to the increase of their local concentration. We analyzed the cross-correlation of the series of impurity concentration smoothed over 10-minute interval with the series of turbulence intensity. Analysis has revealed that the correlation coefficient value is higher than the significance level and equals about 0.3, that is, the turbulence enhances the concentration of suspended particles, most probably due to the transfer.[7] Low correlation can be explained

by a nonlinear character of the relation between the intensity of turbulent processes and the concentration of suspended particles.

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