Spatial structure of mesoscale inhomogeneities in pollutant concentration within the lower troposphere

A.A. Zhuravlev, G.M. Teptin, and O.G. Khutorova

Kazan State University

Received February 7, 2001

Data of many-year minutely measurements of the concentration of aerosol and gaseous pollutants in the near-ground layer have been analyzed to study the spatial structure of mesoscale inhomogeneities in the concentration within the lower troposphere. A periodic structure has been revealed in analyzing longitudinal correlation functions with a varying time lag. Excluding diurnal behavior has allowed us to separate significant peaks in the correlation functions of the concentrations that are most likely caused by transport from neighboring industrial centers. The mean speed of transport ranges from 1.2 to 1.4 m/s, and the mean wind speed exceeds it roughly by 15–20%.

Introduction

In recent six years, the Kazan State University and the Ministry of Nature of the Republic of Tatarstan conduct continuous observations of the concentration of gaseous pollutants and aerosol in the near-ground atmospheric layer at a network of automatic ecological monitoring stations. The stations measure the concentration of the near-ground aerosol and gaseous pollutants (CO, NO, NO_2, SO_2, H_2S), as well as the meteorological parameters (wind velocity, temperature, relative humidity). Measurements are conducted at the altitude of 2.4 m with the interval of 1 min; the results are then averaged over half-hour period. The concentrations of chemical substances are measured accurate to $1\cdot10^{-9}$ g/m^3, and the concentration of aerosol is determined by the filter method with the error of $1\cdot10^{-6}$ g/m^3 (Refs. 1 and 2).

Some results of studying spatial structure of turbulent variations in the concentration of aerosol and some gaseous pollutants have already been published.[3,4] The data obtained at five stations in Almetevsk city allowed us to study the structure of turbulence with the scales ranging from 0.9 to 6.1 km. Analysis was performed by constructing spatial correlation functions of concentrations at the wind direction transverse to the base line. Thus, the effect of transport by the mean wind was excluded.

From this analysis we have revealed stable minimum in the spatial correlation function at the scales ranging from 1.5 to 3.5 km for NO, NO_2, and CO, as well as a weakly pronounced minimum for aerosol. The minimum in spatial correlation of the function of wind azimuth that takes negative values at the inhomogeneity scales ranging from 1.5 to 3.5 km was also observed. At the same time, the correlation function of temperature and relative humidity shows no spatial variations and demonstrates high correlation (0.95–0.99) in the entire range of distances under study. The spatial correlation functions for the opposite wind directions with respect to the base showed insignificant deviations from the mean correlation function.

This is indicative of a significant effect of dynamic processes on the formation of spatial distribution of aerosol and confirms the idea that fluctuations of the concentrations of gaseous pollutants and aerosol in the atmosphere are caused by the effect of turbulence. As it could be expected, vortices with the size of several kilometers do not produce any significant effect on the fields of temperature and humidity. We can state that a vortex structure of 1.5–3.5 km in diameter caused probably by the mesoscale vortices plays a significant part in the scale spectrum of inhomogeneities. Similar results have already been obtained and published by other authors.[5,6]

Experiment

Further analysis of the mesoscale inhomogeneities consists in studying of the correlation functions of the concentrations of gaseous pollutants and aerosol measured by the stations situated in two towns of the Republic of Tatarstan: Almetevsk [53°N, 51°E] – site A and Zelenodolsk [54°N, 49°E] – site B. The distance between these towns (base) is equal to 261 km. Figure 1 shows the map of these towns and several neighboring cities – large industrial centers.

To analyze the data compiled, we have constructed longitudinal correlation time functions with an extended range of time lag variation. First, the series of data was shifted in time by Δt. Then, among measurement data at the site A, we selected those concentrations, when at the moment t_0 the wind direction coincided with the direction of the base and ranged within the following permissible limits (see Fig. 1): $(112 \pm 15)°$ (wind direction from Zelenodolsk to Almetevsk) and $(292 \pm 15)°$ (wind direction from Almetevsk to Zelenodolsk). An additional condition was that at the site B at the time $t_0 + \Delta t$ the wind also

0235-6880/01/06–07 493-04 $02.00

blew along the direction coinciding with the direction of the base and ranging within the permissible interval (for the case of the transport direction from Zelenodolsk to Almetevsk), where Δt is the time lag. The correlation coefficient was determined from the selected data by the equation

$$K[N_{1i}(t_0),\ N_{2i}(t_0 + \Delta t)] = \frac{1}{n} \frac{\sum (N_{1i} - \bar{N}_1)\ (N_{2i} - \bar{N}_2)}{\sigma_{N_1}\ \sigma_{N_2}}, \quad (1)$$

where n is the number of measurements for the given correlation coefficient.

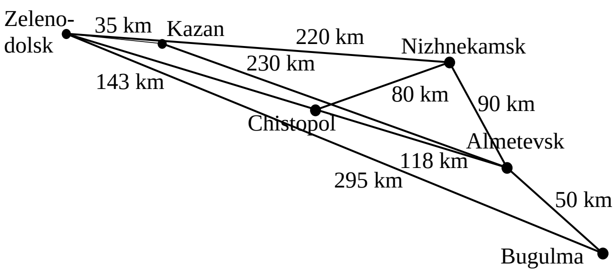


Fig. 1. Geography of towns with the measuring stations and the nearest large industrial centers.

This method allows detection of the processes of pollutant transport in the troposphere by the mean wind and, with the known wind velocity, determination of the distance to the pollution source. To evaluate the reliability of the obtained functions, we have determined the confidence intervals by the following equation[8]:

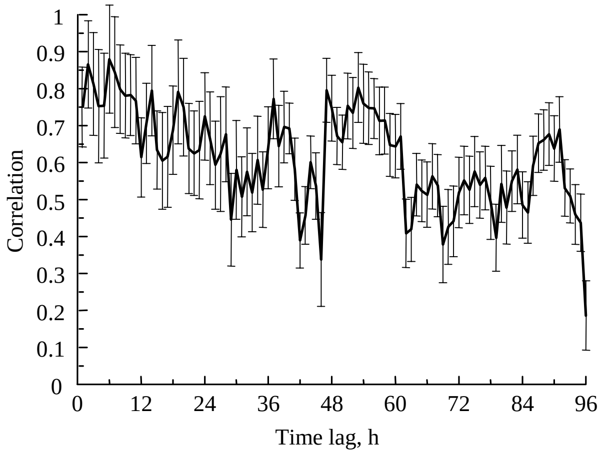
$$\delta_K = \sigma / \sqrt{n-1}\ . \quad (2)$$

Synoptic processes in the atmosphere can conceivably cause the transport of a pollutant within the base. The mean scales of such processes are about several thousands kilometers. Therefore, though the wind inside cyclones and anticyclones has the vortex structure, at the distances of several hundreds kilometers it can be considered codirectional.[7]

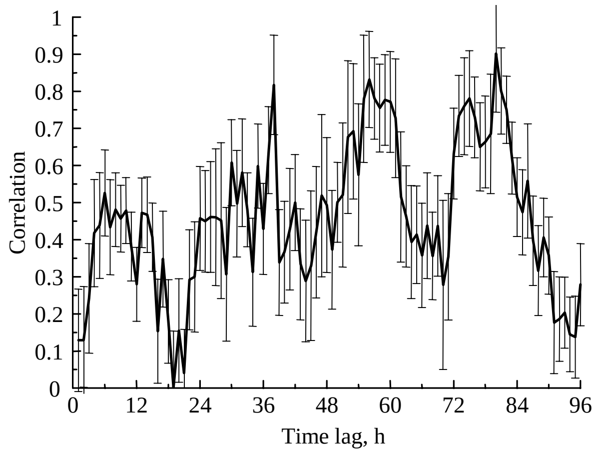
Since three stations in Almetevsk operated simultaneously with one station in Zelenodolsk, we averaged the data of these three stations to exclude local fluctuations of the wind. In addition to the concentrations measured at each station, we took the concentration averaged over the three stations in Almetevsk.

Measurements and calculated results

All the correlation functions obtained have mostly positive values and the shape close to a periodic one (Fig. 2). It is quite likely that various wave processes in the atmosphere cause this. On the average, correlation coefficients vary from 0.4 to 0.8, and the confidence interval ranges from 0.05 to 0.2. Examples of the spatial correlation functions for NO and aerosol are shown in Fig. 2.



a



b

Fig. 2. Longitudinal correlation functions of aerosol (*a*) and NO (*b*) depending on the time shift; wind direction – from Zelenodolsk to Almetevsk.

The plots were analyzed in the following way: (a) distinct, reliable peaks of the correlation functions were separated out, (b) for the time lag corresponding to these peaks, the mean wind velocity was determined, (c) based on the assumption on the existence of pollutant transport, the probable distance to the source of pollution was calculated.

Table 1 presents analysis of the obtained correlation functions for some measured gaseous pollutants and aerosol. The first row gives the time lag, at which the peak in the correlation function is observed, and the second row gives the calculated distance, the pollutant can pass under the effect of the mean wind.

Since the pollutant concentrations vary widely during a day, some peaks in the correlation functions are likely caused by the diurnal and seasonal behavior. The next step of our study was to exclude these variations.

Table 1. Analysis of spatial correlation functions

Substance	Characteristic	Peaks of correlation functions						
NO	Time, h	5–6	12–13	26–27	38–39	52–53	55–58	74–75
	Distance, km	29±2	70±4	170±5	240±8	325±9	350±14	465±18
NO_2	Time, h	6–8	16–19	28	37–39	46–47	58–60	65–67
	Distance, km	45±3	100±5	175±9	230±11	300±12	380±17	395±18
Aerosol	Time, h	6–7	17–18	28	37	47–48	52–56	60–61
	Distance, km	33±2	100±3	175±6	235±8	310±11	360±13	385±17

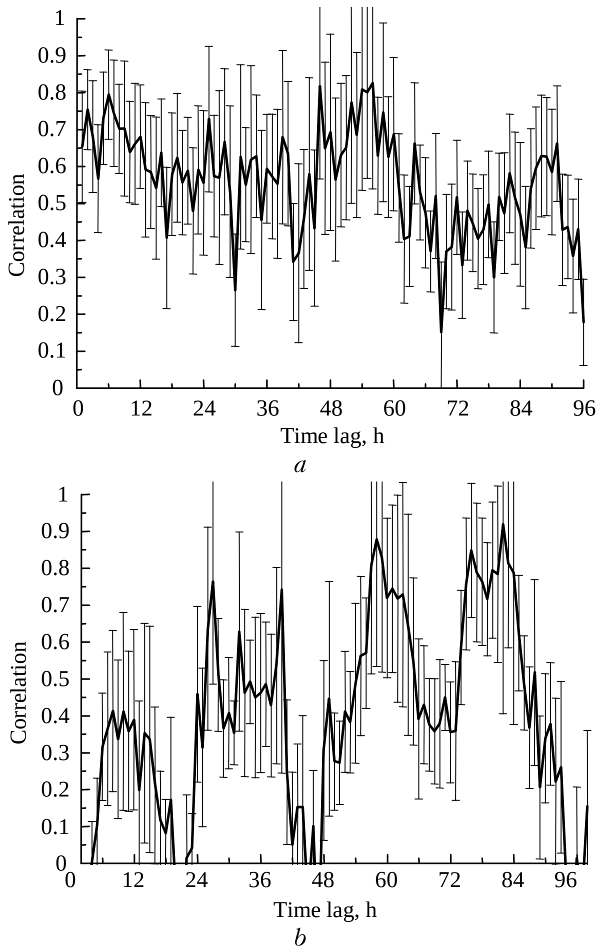


Fig. 3. Correlation function for aerosol (*a*) and NO (*b*) with excluded diurnal behavior; wind direction – from Zelenodolsk to Almetevsk.

It was found that both the shape and the amplitude of the diurnal behavior differ markedly in different seasons. Thus, in summer, the highest concentration is observed between 07:00 and 10:00 L.T. and the lower peak is observed at 21:00 to 22:00 L.T., and from 23:00 until 05:00 L.T. From 12:00 until 18:00 L.T., the concentrations of all the studied substances drop down lower than the diurnal mean value. Both of these peaks are also present in winter, but they are somewhat shifted: the morning peak from 08:00 to 11:00 and the evening one from 16:00 to 19:00, whereas the daytime minimum is weakly pronounced and higher than the diurnally mean value. Besides, the diurnal behavior and the mean concentration depend strongly on the day of a week. This dependence was also taken into account when excluding the diurnal behavior.

For each season and day of a week, the functions of diurnal variability of the concentrations were constructed and then subtracted from the measured values. This allowed us to exclude the diurnal variability more accurately. As an illustration, Fig. 3 shows the same plots as in Fig. 2 but with the subtracted diurnal variability.

The mean level of correlation function becomes somewhat lower, but the height of some peaks even increases a little bit. We applied the 95% confidence interval as the significance criterion.[8]

In the correlation function plotted, one can notice several pronounced statistically significant peaks, which exceed the threshold level determined by the 95% probability. Similar analysis of plots and calculations for statistically reliable peaks of other substances is given in Table 2.

Table 2. Analysis of peaks of correlation functions

Substance		Peaks of correlation			
NO_2	Time, h	6–8	15	38–40	47–48
	Distance, km	40±3	90±4	245±9	285±12
	Site	Sub.	?	Kaz.	Zel.
NO	Time, h	6	25	38	55–57
	Distance, km	35±2	145±4	220±11	310±14
	Site	Sub.	Chis.	?	Zel.
Aerosol	Time, h	5–6	25	46	54–56
	Distance, km	35±5	150±7	280±12	320±15
	Site	Sub.	Chis.	Kaz.	Zel.

Note: The first row gives the time lag, at which a significant peak in the correlation function is observed; the second row gives the calculated distance (km), which the substance could pass under the effect of the mean wind; the third row gives possible source of pollution: Almetevsk suburbs (Sub.), Zelenodolsk, 261 km (Zel.), Chistopol, 114 km (Chis.), Kazan, 230 km (Kaz.), hard-to-explain peaks (?); the distances presented in Note are the actual distances determined based on the geographic map.

Having the diurnal variations excluded, the number of significant peaks for each correlation function decreases a little. The majority of peaks can be explained by pollution transport from neighboring industrial centers. The distance was overestimated somewhat as compared to the actual one that is explained by the known fact that the speed of pollution transport is less than the speed of the mean wind. This is caused by vortices and turbulent inhomogeneities of the atmosphere that decelerate the transport. According to our calculations, the mean speed of transport is, on the average, lower by 15–20%, than the mean wind speed. Thus, the mean wind speed is 1.6–1.9 m/s, and the calculated speed of transport is 1.2–1.4 m/s.

Conclusion

The study of the longitudinal correlation functions with the varying range of the time shift has shown that there are pronounced statistically significant peaks in the correlation function that are partially caused by diurnal and seasonal variations of the concentration. At the same time, analysis shows that some peaks of the correlation function are caused by the transport of substances from neighboring industrial centers. The difference between the wind speed and the transport rate is explained by the theory of turbulent transport and confirms our assumption on the existence of a significant transport of air pollution to the distances up to 300 km. The nature of the periodic variations in the correlation functions is still unclear, but they can be explained by various periodic atmospheric processes. To do this, we should use other methods for analysis, and this is our plan for the future.

Acknowledgments

The work was partially supported by the Russian Foundation for Basic Research, Grant No. 01–0564390, and the Foundation for Research and Experimental Development of the Republic of Tatarstan, Grant No. 09–9.4–52/2001 f.

References

1. A.I. Schepovskih, R.N. Safin, and O.G. Khutorova, Environ. Radioecol. Appl. Ecol. **3**, No. 3, 19–28 (1997).
2. O.G. Khutorova, J. Aerosol Sci. **30**, 335–336 (1999).
3. A.N. Fakhrutdinova and O.G. Khutorova, Izv. Ros. Akad. Nauk, Ser. Fiz. Atmos. Okeana **34**, No. 1, 19–24 (1998).
4. A.A. Jouravlev, O.G. Khutorova, and G.M. Teptin, Environ. Radioecol. Appl. Ecol. **6**, No. 3, 15–19 (2000).
5. G.I. Gorchakov, P.O. Shishkov, V.M. Kopeikin, A.S. Emilenko, A.A. Isakov, P.V. Zakharova, V.N. Sidorov, and K.A. Shukurov, Atmos. Oceanic Opt. **11**, No. 10, 958–962 (1998).
6. G.I. Gorchakov, B.M. Koprov, and K.A. Shukurov, Atmos. Oceanic Opt. **13**, No. 2, 150–153 (2000).
7. A.S. Monin, *Weather Forecast as a Physical Problem* (Nauka, Moscow, 1969), 184 pp.
8. G.M. Jenkins and D.G. Watts, *Spectral Analysis and Its Applications* (Holden-Day, San Francisco, 1968).