Structural changes in aerosol emissions of motor transport in Novosibirsk when changing-over to the lead-free gasoline

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Received November 29, 2000

A few-parameter model is proposed for reconstruction of long-term aerosol pollution of the environment near motorways. The concentrations of lead, polycyclic aromatic hydrocarbons, and macrocomponents in snow are reconstructed using the data of snow sampling late in winter of 1999/2000 in the zone of strong effect of the Sovetskoe motorway (Novosibirsk). Changes in the structure of aerosol emissions of motor transport are analyzed in comparison with the previous winter period.

Introduction

Motor transport is one of the main pollution sources in Novosibirsk. Typical pollutants due to motor car exhausts are sulfur and nitrogen oxides, base metals, in particular, lead, and polyaromatic hydrocarbons (PAH). Emissions of lead and its compounds are especially dangerous for the environment. They are caused by wide application of lead additives (antiknock agents) in production of gasoline with high octane number.

The use of lead-free gasoline is a significant measure decreasing emission of harmful substances, including lead compounds, into the environment. To achieve the expected efficiency, it is necessary to fulfill a number of technical conditions of car engine operation.[1–3] Otherwise, the planned measures could give ambiguous results.

In Novosibirsk, leaded gasoline was intensely replaced by lead-free one in 1999 and 2000. This requires development and application of efficient methods for monitoring of changes. It is worth using snow cover as an indicator of long-term pollution of the environment near motorways.[4,5] To increase the information content of observations, regularities in the spread of pollutants from motorways should be revealed.

1. Reconstruction models

Let the axis y be directed along the motorway, and the axis x be normal to the axis y and lying in the horizontal plane. Then, for a homogeneous terrain, the field $q(x, y)$ of a long-term aerosol pollution near a motorway can be described as follows[6]:

$$q(x,y) = \int_0^{2\pi}\int_{L_1}^{L_2} [S(a)/2\sqrt{\pi k_0 a}]\mathrm{e}^{-b^2/(4k_0 a)} P(\varphi)\mathrm{d}\eta\mathrm{d}\varphi, \quad (1)$$

where L_1 and L_2 are the ends of the motorway section; $a = x\cos\varphi + (y - \eta)\sin\varphi$; $b = -x\sin\varphi + (y - \eta)\cos\varphi$, φ is the angle between the axis x and wind direction; $S(a)$ is the near-surface field of an admixture concentration from a linear source; k_0 is the coefficient characterizing turbulent diffusion of an admixture in the direction normal to the wind; $P(\varphi)$ is the probability of wind direction opposite to φ.

The field of aerosol pollution $S(a)$ from a linear source is determined by solving the semiempiric equation of turbulent diffusion in the atmospheric surface layer[6,7] with natural boundary conditions

$$u\frac{\partial S}{\partial a} - w\frac{\partial S}{\partial z} = \frac{\partial}{\partial z}k(z)\frac{\partial S}{\partial z}, \quad (2)$$

where u is the wind speed in the direction of the movable axis a; w is the sedimentation rate of aerosol; $k(z)$ is the vertical turbulent exchange coefficient.

Equation (1) can be simplified by using the analytical solution of Eq. (2) for the case of exponential representation of the functions $u(z)$ and $k(z)$ in the atmospheric surface layer.[6,7]

In this case, we have the equation[6,8]

$$S(r,\theta) = \frac{\theta_1}{r^{\theta_2}}\exp(-r_\mathrm{m}/r), \quad (3)$$

where

$$\theta_1 = \frac{G\, r_\mathrm{m}^{\omega}}{2(1+n)\, k_1 \Gamma(1+\omega)}; \quad \theta_2 = 1 + \omega;$$

0235-6880/01/03 219-03 $02.00

$$\omega = \frac{w}{k_1(n+1)}; \quad r_m = \frac{u_1 H^{1+n}}{(1+n)^2 k_1}; \qquad (4)$$

G is the capacity of the linear source; n is the exponent in approximation of the wind velocity by an exponential profile; k_1 is the vertical turbulent exchange coefficient at the 1-m height; $\Gamma(1+\omega)$ is the gamma-function.

The use of Eq. (3) allows the number of unknown parameters in Eq. (1) to be decreased down to four ($\theta_1, \theta_2, r_m, k_0$). To determine these parameters, we need measurements of the concentration field at no less than four different points. The number of unknown parameters can be even smaller, if a motorway is normal to the prevailing wind. In this case, the dependence on the parameter k_0 becomes negligible, and the pollution field on the leeward side of the motorway can be reconstructed by Eq. (3) using only estimates of the parameters θ_1 and θ_2. The parameter r_m corresponds to the point of the maximum surface pollutant concentration for a slowly depositing pollutant ($\theta_2 \to 1$) (Ref. 6) and can be predetermined.

2. Estimation of the pollution field

An object of study was a fragment of the Sovetskoe motorway in the left-bank part of the Sovetskii District of Novosibirsk. This fragment of the road is directed from the southeast to the northwest. In winter the right side of the road is subject to the strongest pollution because of high repetition of southern and southwestern winds (more than 60%, Ref. 9). This orientation of the motorway simplifies the study, because it allows us to restrict snow sampling only to the leeward side and use the model (3) for interpretation of observations. Larger-scale study of snow pollution in this region was carried out in late winter of 1999 (Ref. 5). It revealed quantitative regularities in distribution of aerosol pollution. The main goal of the study of 2000 was to refine zones of intense pollution by car exhausts and to estimate characteristics of aerosol emissions. In this connection, snow was sampled at the distance no less than 20 m from the road.

The levels of the snow cover pollution were reconstructed based on the regression dependence of Eq. (3) in the approximation of a heavy and slowly depositing ($\theta_2 \to 1$) admixture. For admixtures with relatively high sedimentation rates, the parameters of Eq. (3) were estimated from the observations obtained at the points spaced by 20 and 35 m from the road (reference points). Other points were used as control points.

Figure 1 shows the reconstructed concentrations of NO_3^-, SO_4^{2-}, benz(a)pyrene, and lead. Analysis of Figs. 1 and 2 shows that the agreement between calculated data and observations at control points is rather good. Some discrepancy is observed at the point spaced by 50 m from the road. This can be explained by insufficient consideration of the effect of polydispersity.

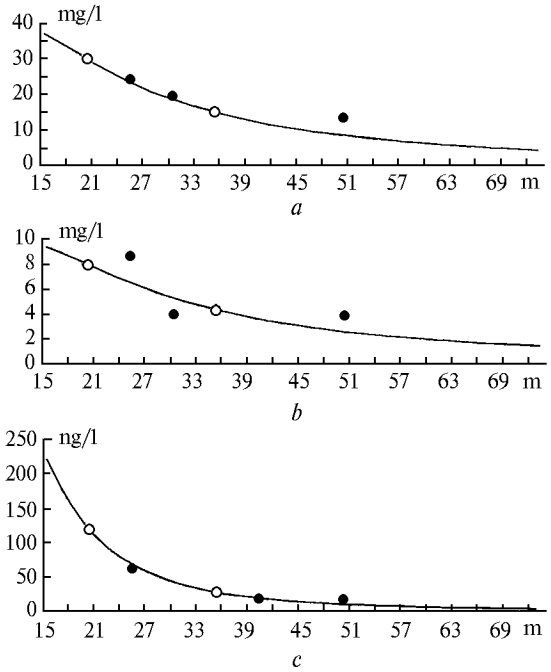

Fig. 1. Calculated and measured concentrations of nitrates (*a*), sulfates (*b*), and benz(a)pyrene (*c*) in snow: reference points (∘) and control points (•).

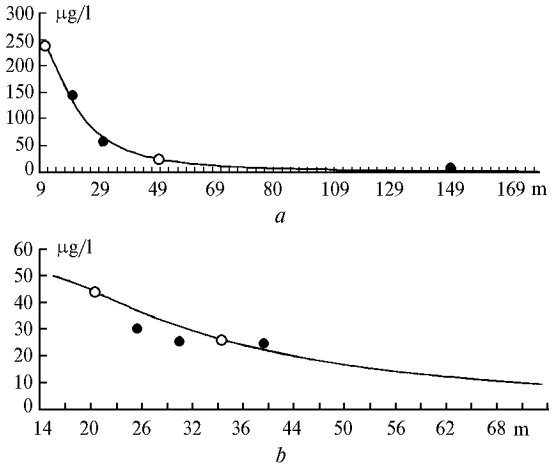

Fig. 2. Distribution of lead in snow for winter seasons of 1998/99 (*a*) and 1999/2000 (*b*).

3. Structural changes in car exhausts

According to Eqs. (3) and (4), the estimates of the parameter θ_2 characterize the mean sedimentation rate w of aerosol particles near the motorway. They are directly connected with the size of aerosol particles. According to Eq. (4), changes in the relative sedimentation rate λ can be described as follows:

$$\lambda = \frac{\theta_2^{**} - 1}{\theta_2^{*} - 1}, \qquad (5)$$

where θ_2^* and θ_2^{**} are estimates of the parameter θ_2 for winter seasons of 1999 and 2000, respectively.

Aerosol fallout M due to car exhausts is estimated based on Eq. (3) by the equation

$$M = \int_{10m}^{50m} S(r, \theta)\, dr. \qquad (6)$$

Table 1 gives the parameters θ_2 and M estimated from the data of snow sampling in 1999 and 2000 for some PAH components. To estimate the total PAH content near the motorway, we invoked the information on moisture content of snow at sampling routes. The mass of snow according to the samples of 1999 was, on the average, 103 kg/m^2; in 2000 it increased up to 140 kg/m^2.

One can see from Table 1 that the content of PAH components in the roadside zone from 10 to 50 m increased from 1.5 to 3 times in 2000. The relative sedimentation rate of aerosol particles containing PAH changed significantly. Its growth for different PAH components was from 4 to 10 times. It follows herefrom that the mean size of particles with PAH increased, according to the Stokes equation, from 2 to 3 times.

Table 1. Estimates of total fallout and relative sedimentation rate of PAH components

PAH	θ_2		Fallout M, g/km		Relative sedimentation rate, λ
	1999	2000	1999	2000	
Benz(a)pyrene	1.27	3.6	0.16	0.55	9.6
Fluoranthene	1.45	4.08	1.2	1.9	7
Pyrene	1.72	4.2	0.6	1.5	4.4

Table 2 gives the data on the distribution of lead in different forms for winter seasons of 1998/99 and 1999/2000. The analysis of this table shows that the distribution of lead between fractions changed significantly by 2000. If in 1999 the coarse fraction dominated absolutely for all distances, then in 2000 the total contribution of the fine fraction and water-soluble part became comparable with it. For the roadside zone from 10 to 50 m, the total lead content in snow for winter of 1998/1999 was about 400 g/km, and in the winter season of 1999/2000 it was roughly 193 g/km. From Table 2 it follows that the decrease in the lead content is connected largely with the decrease of the coarse component.

Table 2. Fraction distribution of lead, %

Winter season	Fraction	Distance to motorway, m		
		20	30	50
1998/99	A	1.4	2.7	2.8
	F	0.4	6.1	2.5
	C	98.2	91.2	94.7
1999/2000	A	7	14	9
	F	22	28	30
	C	71	59	61

Note. A – aqueous, F – fine, and C – coarse fractions.

Conclusion

This study allows us to draw some conclusions.

In the zone of intense pollution by car exhausts, the distributions of concentrations of macrocomponents and PAH, as well as the total content of lead in soluble and insoluble fractions are well described by the model (3) of a linear elevated source. The surface concentration for a slowly depositing admixture is maximum at the distance about 25 m from the road.

The largest changes in the winter season of 1999/2000 are observed in emissions of PAH. The total content in the roadside zone from 10 to 50 m increased 3 times for benz(a)pyrene, and 1.5 and 2.5 times for fluoranthene and pyrene, respectively. The mean size of aerosol particles containing PAH also increased markedly. For the PAH components under consideration, it changes from 2 to 3 times as compared to the winter season of 1998/99.

Due to the use of large amounts of lead-free gasoline along with the leaded one, significant changes are observed both in the structure of aerosol emissions of motor transport and in the mass of emitted pollutants. The total content of lead decreased in 2000 mostly at the expense of the coarse fraction. This circumstance should be taken into account when evaluating the efficiency of the taken measures, since the coarse fraction of lead is less dangerous for the environment.

To draw a more complete pattern of pollution of the roadside zone, additional studies are needed. They should take into consideration the effect of polydispersity and the influence of snowploughs.

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