# Control of Aerosol Emission Near Motorways

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## Abstract

A model of long-term pollution of environment by motor transport emission is proposed. The numerical model was tested for the recovery of long-term aerosol pollution of the locality by polycyclic aromatic hydrocarbons, lead, macrocomponents; the data of strip snow surveys near the Barnaul motorway and the Sovetskoye highway in Novosibirsk are involved. The structural changes of aerosol emission of motor transport during winter seasons of 1998–2001 are analyzed.

## INTRODUCTION

Motor transport running on Diesel fuel and on ethylated petrol is one of the basic sources of environmental pollution [1]. Characteristic pollutants are sulphur and nitrogen oxides, as well as their transformation products sulphuric and nitric acids, heavy metals, in particular lead, polycyclic aromatic hydrocarbons (PAHs). These substances pass into the environment mainly with exhaust gases. As a consequence, the highest contribution into pollution is made by motorways.

At the same time, investigations related to this problem focus mainly on the study of the consequences of this pollution for agricultural plants [2, 3], on the determination of soil pollution with heavy metals. A characteristic feature of these investigations is labour-intensiveness connected with sampling and chemical analysis of the component composition of samples. As far as the investigation of air pollution is concerned, it is performed periodically by monitoring services of Goskomgidromet (State Committee for Hydrometeorology) and Goskomsanepidnadzor (State Committee for Sanitary and Epidemiological Inspection) [4–7].

In order to elaborate measures for the protection of the atmosphere from pollution, it is necessary to involve both chemical analytical methods and the means of mathematical geophysics for estimating emission from motorways. The emission depends on a large number of parameters: characteristic categories, intensity and velocities of automobiles, wear and operation level of motors, fuel type, *etc*. A series of procedures has been developed to evaluate the gas constituents of the emission. The data obtained allow one to analyze the pollution status of the atmosphere both over

the entire cities and over separate motorways [5–9].

Long-term pollution, especially aerosol one, in the vicinity of motorways has been much poorer investigated, both in the aspect of experimental investigation and in quantitative description. The most promising object of investigation is snow cover [10–12]. The use of snow cover as an indicator of the level of aerosol pollution of environment provides unique possibilities for efficient monitoring of automobile emission under the conditions of Siberia. Relatively simple snow sampling procedure allows one to perform a large-scale examination of the vicinity of motorways.

On the basis of the data on the chemical composition of snow, involving theoretical notions on aerosol admixtures spreading in the surface layer of the atmosphere, we investigated the spatial regularities of aerosol deposition of dust, heavy metals, in particular lead, PAHs, macrocomponents. We estimated the characteristics of aerosol emission from motors in Novosibirsk for a series of winter seasons with the help of the obtained regularities.

## EXPERIMENTAL CONDITIONS AND OBSERVATION PLANNING

A systematic investigation of snow cover pollution was performed in the vicinity of the Barnaul motorway and near the Sovetskoye highway in Novosibirsk. Snow was sampled at the end of winter seasons 1998–2001; this allowed us to follow the dynamics of long-term pollution, changes in the component and disperse composition, and to estimate total emission of motor transport [10, 11]. The regions of both motorways near which the surveys were performed are similarly directed passing from south-east to north-west; the distance between them is about 30 km. The land surface near the motorways is uniform and practically devoid of plants. This allows us to perform direct comparison between the obtained results and to simplify substantially the interpretation of the observation data, since wind direction recurrence is more than 60 % for the southern and south-western wind directions. The orientation of the chosen parts of motorways

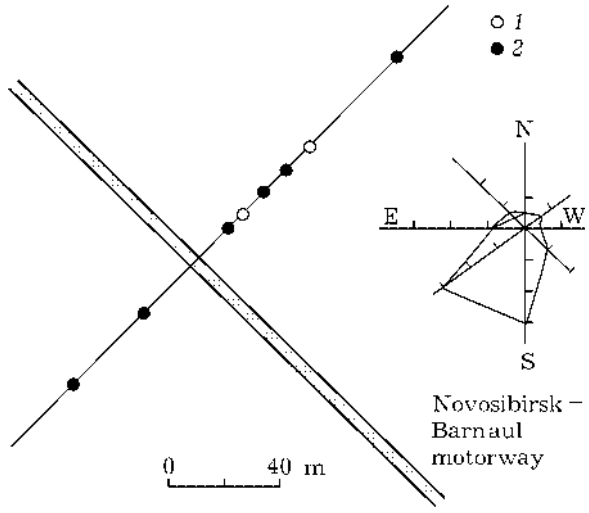


Fig. 1. Scheme of the snow survey: *1* – reference points, *2* – control points.

points to the necessity of more detailed investigation of the north-eastern side of the roads.

Snow cover was sampled over its whole height with the help of a tube made of titanium (∅ 100 mm), preventing the capture of soil particles from the land surface with the help of a polyethylene spade. As a rule, sampling sites were located at both sides of the road within 10 to 150 m; the points were situated more dense at the nearest end of the section and gradually they became more rare with an increase in the distance from the road. This was due to different intensities of the aerosol pollution processes in the nearby and remote regions. The territory directly adjacent to the road is affected by snow-ploughs, which requires separate analysis. A typical layout of sampling points is shown in Fig. 1.

For numerical modeling of the optimal location of sampling sites, it is necessary to possess rather adequate model of the processes involved in long-term pollution of the territory, as well as *a priori* information on model parameters [13, 14]. In the absence of the data on the necessary parameters, preliminary experimental investigation is required.

## MODEL OF LONG-TERM POLLUTION RECOVERY

Let us consider a linear source of finite length located at the $y$ axis within the interval $(L_1, L_2)$. Let the horizontal wind direction form

angle β with the $x$ axis (measuring from the $x$ axis counter-clockwise). Spreading of the gas admixture from a point source located at a point $\eta \in (L_1, L_2)$ can be described by the following equation:

$$q = \frac{M}{(1+n)k_1\varphi_0 x_P^2\sqrt{2\pi}} \times \exp\left(-\frac{u_1 H^{1+n}}{k_1(1+n)^2 x_P} - \frac{y_P^2}{2\varphi_0^2 x_P^2}\right) \quad (1)$$

where $M$ is the source power; $k_1$, $u_1$ are the coefficient of vertical turbulent exchange and the wind velocity at a height of 1 m, respectively; $\varphi_0$ is the dispersion of deviations in the directions of wind velocities; $n$ is the power in the equation approximating wind velocity by a power profile; $H$ is the effective height of the source; $x_P = a - \eta\sin\beta$, $y_P = b - \eta\cos\beta$, $a = x\cos\beta + y\sin\beta$, $b = -x\sin\beta + y\cos\beta$.

Then, taking into account (1), we can represent the equation for calculating long-term aerosol pollution of the vicinity of motorway as follows [11]:

$$S(x, y, \Theta_0, \Theta_1) = \Theta_0 \int_0^{2\pi}\int_{L_1}^{\eta_p} q_n(x_P, y_P, \alpha, \varphi_0) \times \lambda(x_P, \Theta_1) R(\varphi + 180°) d\eta d\varphi \quad (2)$$

where

$$\Theta_0 = \frac{M}{(1+n)k_1\varphi_0\sqrt{2\pi}}, \quad q_n = x_P^{-2} e^{-\frac{\alpha}{x_P} - \frac{y_P^2}{2\varphi_0^2 x_P^2}}$$

$$\alpha = \frac{u_1 H^{1+n}}{(1+n)^2 k_1}$$

$$\eta_P = \begin{cases} L_2 \text{ at } y + x\,\mathrm{ctg}\,\beta \geq L_2 \\ y + x\,\mathrm{ctg}\,\beta \text{ in the opposite case} \end{cases}$$

$$\lambda(x_P, \Theta_1) = x_P^{\Theta_1}, \quad \Theta_1 = -\frac{w}{k_1(n+1)} \quad (3)$$

$w$ is the mean rate of aerosol particle deposition, $R(\varphi)$ is the winter mean wind rose for the territory under consideration. The $R(\varphi)$ function is usually measured for a discrete set of angles $\varphi_j = j\Delta\varphi / N$, $j = 1, \ldots, N$, where $\Delta\varphi_j = 2\pi$, $N$ is the number of compass points. In order to obtain continuous dependence of the $R$ function on $\Delta\varphi$, it is useful to apply the following linear interpolation:

$$R_j(\varphi) = P_j + \frac{(P_{j+1} - P_j)\varphi}{\Delta\varphi}, \quad j\Delta\varphi \leq \varphi \leq (j+1)\Delta\varphi$$

Here $P_j$ is the recurrence of wind direction for a $j$-th compass point, $j = \overline{I, N}$.

Important parameters in eq. (2) are $\Theta_0$, $\Theta_1$. In order to estimate them, one should perform observations in at least two points of the territory. The estimation procedure can be substantially simplified if we take into account the linearity of eq. (2) with respect to the parameter $\Theta_0$. This allows us to perform separate determination of the parameters $\Theta_0$ and $\Theta_1$. In order to improve the stability of estimation, it is reasonable to use optimal sampling schemes at the stage of experimental investigation. The calculation of optimal positions of sampling sites can be made on the basis of the algorithms of consequent analysis and planning of experiment [13, 14].

In the case under consideration, in order to estimate long-term aerosol pollution, the direction of the selected regions of motorways near which sampling was performed allows us to use both the full model (2) and an approximate one [10]

$$S(x, \Theta_0, \Theta_1) = \frac{\Theta_0}{x^{1+\Theta_1}} \exp\left(-\frac{\alpha}{x}\right) \quad (4)$$

Model (2) allows recovering the fields of concentrations of pollutants in snow at the same time at both sides of motorway. In this case, the information of wind rose for the given winter season is necessary. When (4) is used, no detailed information on wind rose is required.

## CHEMICAL ANALYSIS OF SNOW SAMPLES

After transferring the samples into the liquid state, we used 2 analysis schemes depending on the nature of components to be analyzed. For the analysis of inorganic components, the melted sample was first filtered though a paper filter (blue ribbon), then through a membrane filter (∅ 0.45 μm). The resulting precipitates were dried in the air; the precipitates and the filtrates were analyzed. The determination of PAHs was performed after preliminary extractive preconcentrating in hexane from the whole volume

TABLE 1

Components under determination in snow cover samples, and the analysis procedures

| Components | Analysis procedures |
|---|---|
| pH, $HCO_3^-$ | Potentiometry (Anion-410) |
| $SO_4^{2-}$, $Cl^-$, $NO_3^-$ | Ion chromatography (Waters, USA) |
| Ca, Mg, Na, Zn, K | Atomic absorption flame spectrometry (Hitachi 8000, Japan) |
| Ba, Fe, Cd, Mn, Cu, Pb, Cr, Ni, Be | Atomic emission spectroscopy with arc excitation of spectra (PGS-2, Germany) |
| $NH_4^+$ | Photocolorimetry (SQ-118, Merck, Germany) |
| PAH | Chromatomass spectrometry (Hewlett Packard, Germany) |

of non-filtered sample [11]. The components under determination are listed in Table 1.

The analysis was performed in the analytical laboratories of the Institute of Inorganic Chemistry and Institute of Organic Chemistry, SB RAS, accredited at the Gosstandart (State Committee of Standards) of RF, and in the SSC "Vector". Analysis errors were within the limits defined by the State Standard 27384–87.

## ESTIMATION OF CHARACTERISTICS OF AEROSOL DEPOSITION

For the qualitative and quantitative analysis of aerosol deposition processes in the vicinity of motorways, let us limit our consideration to the most representative experimental data obtained in winter seasons 1998–2001.

### *Macrocomponents*

The data of experimental investigation of the macrocomponent composition of snow samples collected in the end of February, 2001 near the Sovetskoye highway are shown in Table 2. It follows from these data that in general a monotonous decrease in the content of all the macrocomponents is observed while the distance from the road increases. These components are mainly constituents of the road dust, so their maximal content is observed near the road and decreases with increasing distance [10]. In order to describe their distribution, it is reasonable to use model (4).

It should be noted that macrocomponents of the water filtrate are transferred mainly in the coarse fraction of the road dust. The level

TABLE 2

Spatial dynamics of specific content of macrocomponents, mg/l (Sovetskoye highway, 2001)

| Component | Distance from the road, m | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | North-East | | | | | | South-West | |
| | 15 | 20 | 30 | 40 | 50 | 70 | 35 | 50 |
| $Ca^{2+}$ | 6.09 | 5.56 | 4.46 | 4.91 | 3.88 | 3.16 | 2.68 | 2.17 |
| $Mg^{2+}$ | 0.50 | 0.54 | 0.30 | 0.31 | 0.27 | 0.21 | 0.23 | 0.24 |
| $Na^+$ | 2.66 | 2.26 | 0.86 | 0.54 | 0.47 | 0.38 | 0.25 | 0.40 |
| $K^+$ | 0.38 | 0.30 | 0.22 | 0.23 | 0.21 | 0.26 | 0.13 | 0.20 |
| $HCO_3^-$ | 17.4 | 22.5 | 14.1 | 16.5 | 14.8 | 10.4 | 10.5 | 6.8 |
| $NO_3^-$ | 10.2 | 4.0 | 2.1 | 2.1 | 1.3 | 1.5 | 1.1 | 0.80 |
| $SO_4^{2-}$ | 23 | 7 | 7 | 8 | 6 | 7 | <6 | 6 |
| $Cl^-$ | 3.23 | 1.57 | 0.36 | 0.29 | 0.29 | 0.29 | 0.14 | 0.14 |
| æ$^{18\,°C}$, μS/cm | 55.5 | 52.6 | 30.6 | 36.4 | 23.9 | 18.2 | 18.2 | 14.4 |

*Note.* Relative standard deviation ($S_r$) of the procedures of Ca, Mg, Na, K determination was 0.03–0.05; $HCO_3^-$ 0.04–0.06; $Cl^-$ 0.05–0.10; $SO_4^{2-}$ 0.07–0.12, specific electric conductivity 0.07–0.10.

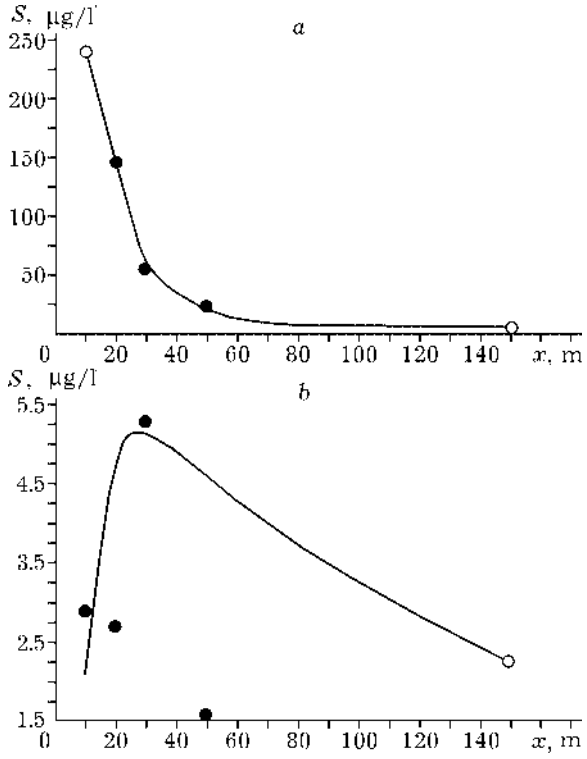


Fig. 2. Specific lead content in the coarse part (*a*) and in the sum of fine and water-soluble parts (*b*).

of dust component content near motorways is due to snow ploughs and wind transport from the road surface. The dynamic and thermal action of the moving vehicles can provide additional elevation of fine fractions of dust at some height and thus lead to their deposition in the direction of prevailing winds at a distance of 20–30 m from the road surface. The ratio of components in the coarse and fine fractions determines the dynamics of their distribution with an increase in the distance from the road.

### *Lead*

The results of estimations of lead content in snow for winter 1998–1999 near the Sovetskoye highway with the model (4) are shown in Fig. 2 [10]. The recovery of spatial distribution of the coarse fraction of lead is shown in Fig. 2, *a*; the results of numerical modeling of the total lead content of fine and water-soluble fractions are shown in Fig. 2, *b*. The model describing the expansion of weakly precipitating admixture corresponds to $\Theta_1 \approx 0$. In this case, in order to estimate the parameter $\Theta_0$ of regression function (4), only one observation site was involved, which was located at a distance of 150 m from the road.

The dynamics of the distribution of lead over fractions for the recent three winter seasons is shown in Fig. 3. One can see that the trend of increasing relative content of fine and water-soluble fractions is observed, which is due to the transfer of motor transport from ethylated petrol to non-ethylated one.

Within the winter season 2000–2001, total accumulation of lead in the strip at a distance of 10–50 m from the Sovetskoye highway was about 140 g/km. This is somewhat lower than the value for the winter season 1999–2000 (190 g/km). The estimated amount of total emission of lead during the winter 1998–1999 was 400 g/km [11].

### *Polycyclic aromatic hydrocarbons*

An estimation of concentration fields of benz(a)pyrene (BP) in snow was performed using the data of experimental investigations near the Sovetskoye highway and the Barnaul motorway with the help of regression dependence (2). The reference observation sites were selected to the north-east from the roads at a distance of 20 and 50 m. The results of the recovery of the fields of aerosol deposition of

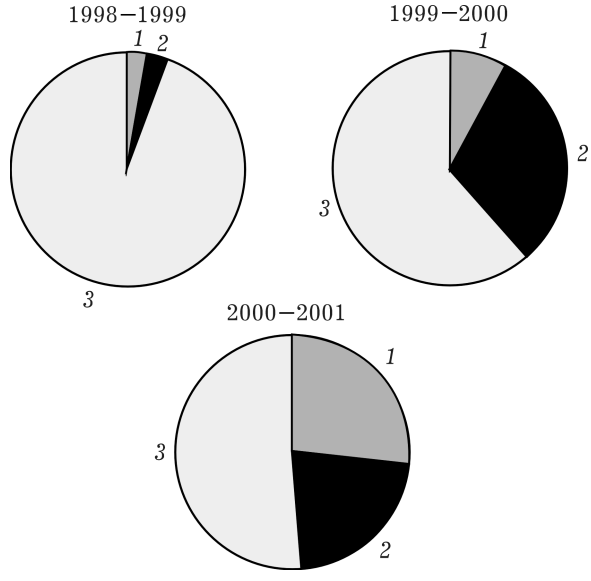


Fig. 3. Distribution of lead over fractions at a distance of 50 m from the road in winter seasons of 1998–2001: *1* – water-soluble fraction, *2* – fine fraction, *3* – coarse fraction.

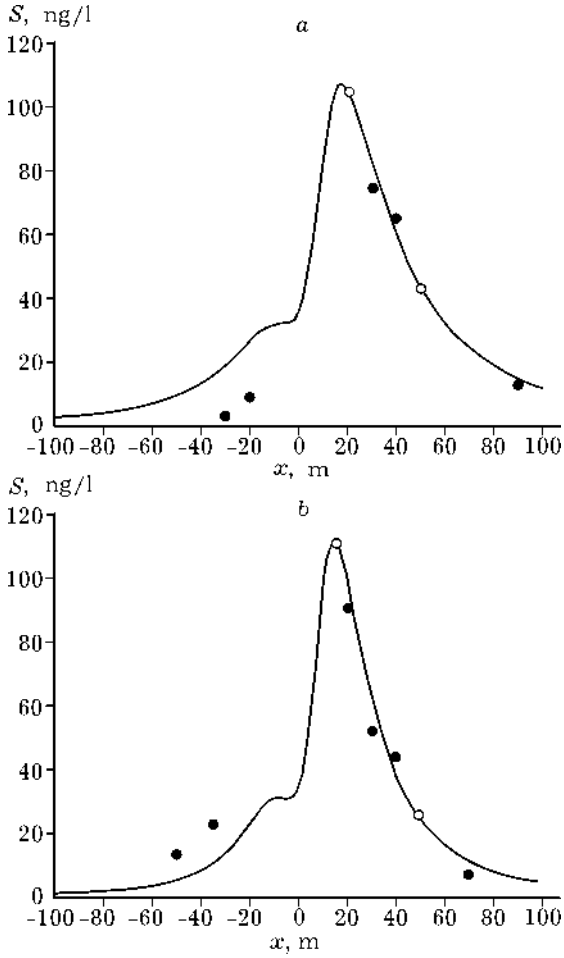


Fig. 4. Calculated and measured specific content of benz(a) pyrene in snow or the Barnaul motorway (*a*) and the Sovetskoye highway (*b*).

BP are shown in Fig. 4. The data on estimations of the parameters for various PAH components are shown in Table 3 [10]. The analysis of the presented information demonstrates rather good consistency between calculation results and observation. The obtained curves exhibit similarity, which is explained by coincidence of estimations of $\Theta_1$ parameters for both sets of experimental data.

Estimations of $\Theta_0$ parameters are linearly linked with the source power. Comparison of $\Theta_0$ estimations shows that the emissions of BP for both roads are quite comparable. In the nearest region, the BP content of snow is about 100 ng/l, which can cause a noticeable negative effect on the health (maximum permissible concentration for water is 5 ng/l).

The $\Theta_1$ parameters for winter season 2000–2001 are somewhat lower than those for the winter season 1999–2000. In consistency with (3), this means that the average size of aerosol particles containing PAHs has correspondingly decreased. The total emission of PAHs during these winter seasons was practically the same; it remained rather high in comparison with the winter season 1998–1999 [11].

## CONCLUSIONS

A complex investigation of the pollution of snow cover by motor transport emissions allows us to make the following conclusions.

The proposed numerical model of long-term pollution of the territory by motor transport emission provides the possibility to perform both the recovery of concentrations of aerosol admixtures at both sides of the roads, and their recalculation into arbitrary orientation of a road, which broadens the possibilities of interpreting experimental studies.

The test of the proposed model demonstrated its satisfactory consistency with the data of observations of PAHs, different fractions

TABLE 3

Measured (numerator) and recovered (denominator) PAH concentrations near the Sovetskoye highway (the data of the season 1998–1999), ng/l

| PAH | Diastance from the road, m | | | | | | Estimations of parameters | |
|---|---|---|---|---|---|---|---|---|
| | 10 | 20* | 30 | 50* | 75 | 100 | $Q_1/10^3$ | $Q_2$ |
| Benz(a)pyrene | 209/29 | 47 | 45/43.2 | 31 | 16/20.9 | 16/15.2 | 16.3 | 1.45 |
| Fluoranthene | 1314/250 | 370 | 334/319 | 210 | 127/143 | 74/94 | 200 | 1.6 |
| Pyrene | 842/163 | 196 | 150/149.7 | 85 | 51/48 | 35/31 | 260 | 1.9 |

*Note.* The sites marked with asterisks were used to estimate regression parameters (4). Only the measured PAH content values are listed for them. The $S_r$ of determination procedures was 0.10–0.25.

of lead, and macrocomponents. The comparison of the PAH profiles recovered from measurement data for the Sovetskoye highway and the Barnaul motorway demonstrated that they were almost identical. Rather high level of PAH content in snow in the nearest region (20–40 m from the road) should be stressed.

Substantial structural changes in aerosol emission from motor transport, due to its transfer for the use of non-ethylated petrol, are confirmed by the analysis of lead and PAH content in snow on the basis of the data obtained during the three winter seasons (1999–2000). Taking into account the extent of the environmental effect of motor transport emission, it is necessary to develop theoretical and applied research of this problem, to create optimal and economic schemes of monitoring, data analysis, and to elaborate efficient measures to decrease the negative action.

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