Study of aerosol fallout of polycyclic aromatic hydrocarbons near the Novosibirsk electrode-producing plant

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Received September 2, 2000

The specific content of polycyclic aromatic hydrocarbons in the vicinity of the plant is reconstructed from the data of snow sampling at a limited number of points. Comparison of calculated and experimental data demonstrates their close agreement. Using the data on climatic repetition of wind velocities in winter, a model of kinematic pollution of the territory is proposed. The model was used for estimation of rates of polycyclic aromatic hydrocarbons fallout.

Introduction

The Novosibirsk electrode-producing plant situated 80 km to the south from Novosibirsk produces graphite electrodes and carbons and significantly contributes to environmental pollution by polycyclic aromatic hydrocarbons (PAH), including benzapilene. To determine the PAH fallout zones, in 1993–1994 the territory surrounding the plant was surveyed at the area more than 100 km^2. As the result, a patchy hard-to-interpret pattern of pollution was revealed. In this connection, in 1995 the Vektor–Ekotsentr organization started regular sampling of snow in the vicinity of the plant, which allowed one to reconstruct the benzapilene pollution fields with sufficiently high accuracy, as well as estimate its total emission for the winter period.[1]

In this paper, we continue the research presented in Ref. 1 using the data of snow sampling late in winter of 1999 in the plant influence zone. Along with benzapilene, the contents of other PAH in snow were measured. The data of chemical analyses were quantitatively interpreted based on the solution of the semiempirical equation of turbulent diffusion using a small number of sampling points. This allowed the data obtained at the rest of the points to be used for estimating the accuracy of reconstruction of pollution fields for different PAH. Estimation of model parameters revealed the similar, on the whole, character of the PAH content change with the distance from the source.

Arrangement of zones of maximum pollution and variation of the concentration with the distance from the plant suggest that the portion of fine particles containing PAH is relatively small in this case. The main amount of PAH comes to the atmosphere with large composite particles due to peculiarities of the technology. In this connection, the kinematic model of PAH pollution in the near zone is considered.[2,3] In this paper, the mean rate of PAH aerosol sedimentation in the atmosphere is estimated based on the climatic data on repetition of wind velocities in winter and PAH concentration fields reconstructed from the observational data.

1. Experimental study of snow cover pollution

Sampling points should be arranged based on the pre-analysis of the data on PAH sources, local conditions, system of roads, housing, and forestation, state of the snow cover, climatic characteristics of wind repetition and velocity in the winter period, and so on.

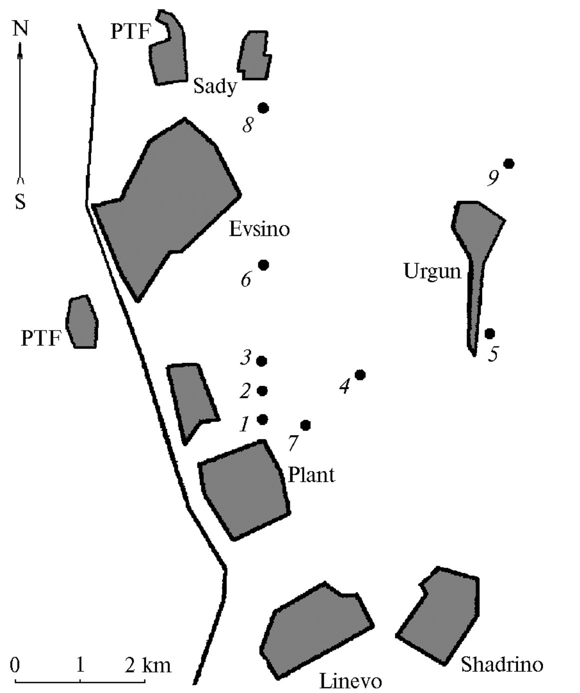

Fig. 1. Arrangement of snow sampling points at the territory.

According to Ref. 4, the major portion of PAH is emitted from two neighboring smoke stacks of a calcining shop. Taking into account all the aforesaid, the flat territory and the absence of other PAH sources in the direction of dominant winds allow the route snow sampling to be chosen as the main research method.[5,6]

Figure 1 shows the position of points at the routes in the northern and northeastern directions. The pre-analysis of experimental data of soil pollution by PAH near the plant[4] with allowance made for the findings from Refs. 1 and 7 allowed us to optimize the arrangement of the sampling points (points No. 1 and No. 3). The use of two routes improves the accuracy of estimation of snow cover pollution fields. Comparing the reconstructed and measured concentrations, we can estimate the extra income of PAH from villages Evsino and Urgun to the points No. 8 and No. 9, respectively.

Table 1 presents the main characteristics of sampling points and the data of chemical analysis.

2. Reconstruction of snow pollution from experimental findings

To describe quantitatively the long process of deposition of PAH emitted from smoke stacks along the route, we use the regression dependence proposed in Refs. 1 and 6:

$$p(r, \boldsymbol{\theta}) = \theta_1 g(r, \theta_2) f(r), \tag{1}$$

where $p(r, \boldsymbol{\theta})$ is the specific content of aerosol impurity in snow; r is the distance from the source; $\boldsymbol{\theta} = (\theta_1, \theta_2)$ is the vector of unknown parameters;

$$g(r, \theta_2) = r^{-\theta_2}; \quad \theta_2 > 0;$$

$$f(r) = \frac{1}{r^2} e^{-2r_{max}/r}, \tag{2}$$

where r_{max} is the distance from the source, at which the surface concentration of a weightless impurity emitted by this source is maximum.

The value of r_{max} is determined by the source geometry (stack height and diameter), parameters of the emitted gas–air mixture, mean winter wind velocity, and air temperature. In our case, $r_{max} \approx 3$ km.

The function $f(r)$ describes relative distribution of surface concentration of the weightless impurity. The function $g(r, \theta_2)$ corrects taking account of the sedimentation processes for heavy impurities. The parameter θ_1 is proportional to the emission power and depends on the climatic characteristics (wind, turbulent exchange coefficients, and sedimentation rate of aerosol impurity) in a rather complex way.[1] The parameter θ_2 is complex too:

$$\theta_2 = \frac{w}{k_1(n+1)}, \tag{3}$$

where w is the sedimentation rate of aerosol particles; k_1 is the coefficient of vertical turbulent exchange; n is the exponent in the approximation of the horizontal wind velocity component by power profile.

The difference in concentrations of light and heavy impurities is mostly determined by the dimensionless parameter θ_2 (Ref. 8).

To estimate the parameters of the regression (1), we have to use observations at two points. Their optimal arrangement can be found using the results from Ref. 9. Then it can be presented as

$$r_{1,2}(\theta_2) = \frac{2\,r_{max}}{\theta_2 + 2}\left(1 \pm \frac{1}{\sqrt{5 + 2\theta_2}}\right). \tag{4}$$

Since the optimal position of the points (4) depends on θ_2, it is necessary to have *a priori* information on this parameter or conduct preliminary observations for its estimation and further refinement.[9]

Table 1. PAH concentration (μg/l) near the Novosibirsk electrode-producing plant (measured values are in numerator and calculated ones are in denominator)

PAH	Point numbers					Estimates	
	1*	2	3*	4	5	$\theta_1 \cdot 10^{-3}$	θ_2
Fluorene	339/339	272/421	267/267	121/142	7.1/10.4	188	3.84
Pyrene	451/451	322/449	254/254	145/121	9.0/6.5	221	4.28
Benz(a)anthracene	2427/2427	1850/2712	1610/1610	865/824	61/52.3	1268	4.04
Perilene	1571/1571	1139/1877	1165/1165	605/607	41.80/41.84	854	3.92
Benz(a)fluorant	454/454	311/353	173/173	117/74	10.3/2.8	194	4.76
Benzapilene	27.8/27.8	18.4/28.7	16.4/16.4	6.9/8.03	0.40/0.46	13.9	4.2
Distance, km	0.75	1.25	1.65	2.4	4.2		
Direction	N	N	N	NE	NE		

Note. Asterisks denote the points used for estimation of the regression parameters (1).

The estimate $\theta_2 \approx 4$ was found[1] from the data on benzapilene content in soil. In this case, according to Eq. (4), the points r_1 and r_2 should be roughly 0.8 and 1.3 km, respectively, far from the smoke stacks.

The parameters θ_1 and θ_2 estimated for different PAH components are presented in Table 1. Table 1 and Fig. 2 give the PAH concentrations in snow, reconstructed from the data obtained for the two observational points. The points in Fig. 2 are marked by circles. The rest of the points can be used to estimate the accuracy of the reconstruction.

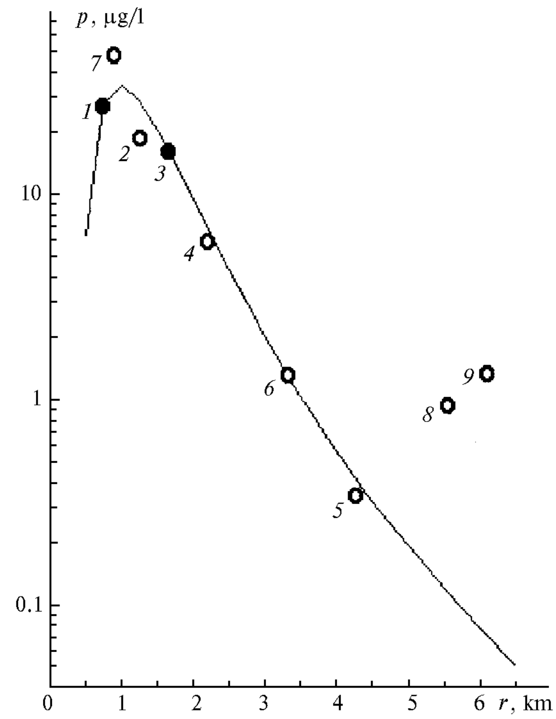

Fig. 2. Specific concentration of benzapilene (in µg/l): the curve shows the result of reconstruction; the reference points are marked by closed circles, and the check points are marked by open circles.

The deviations between the calculated and measured PAH concentrations at the points No. 8 and No. 9 are indicative of significant contribution in the pollution of heating systems in villages Evsino and Urgun, which use local coal with high PAH content.

3. Estimation of rates of aerosol PAH fallout

The contrast change of the PAH concentration in snow with the distance from the source and very close position of the surface concentration maximum are indicative of the high sedimentation rate of PAH contained in coarse aerosol particles. At such rates, the turbulent exchange plays an insignificant role in PAH transport onto the surface,[2,3] whereas the mechanisms of kinematic distribution are prevailing. In this case, the PAH concentration with the distance from the source is determined mostly by the climatic variability of the wind velocity in the winter period.

Table 2 gives the data on climatic distribution of the repetition of wind velocity for Novosibirsk in the cold season at the altitude of 200 m for five gradations of the wind velocity, which cover 99.6% of all cases.[10] For each velocity gradation u_i, $i = 1, \ldots, 5$, the rate of gravitational sedimentation w_i of aerosol particles is determined as

$$w_i = \frac{(H + \Delta H_i)u_{i,\mathrm{m}}}{r_i},\ i = 1,\ \ldots,\ 5\ , \qquad (5)$$

where H is the height of smoke stacks (180 m in our case); ΔH_i is the extra height of the aerosol lift due to plume buoyancy and dynamic effects[8]; $u_{i,\mathrm{m}}$ is the mean wind velocity in the layer of PAH particles sedimentation; r_i is the distance from the source, within which the PAH particles of the ith gradation are deposited.

Table 2. Characteristics of benzapilene (BP) sedimentation at five gradations of wind velocity

Gradation of wind speed, m/s	Repetition, %	Sedimentation rate, cm/s	u_m, m/s	Maximum distance of BP carry-over, m
0–4.5	18.2	85	3.5	825
0–9.5	58	98	7.4	1350
0–14.5	86.6	96	11.2	2100
0–19.5	97.5	85	15.1	3200
0–24.5	99.6	84	19	4050

Based on the regression dependence (1) and the data of Table 1, r_i is determined from the following equations:

$$\Phi(r_i)/\Phi(R) = \lambda_i,\ i = 1,\ \ldots\ ,\ 5\ , \qquad (6)$$

where

$$\Phi(r) = \int_0^r p(s,\ \theta)\mathrm{d}s;$$

R is the distance from the source, within which the emitted BP aerosol is fully deposited; λ_i is the repetition of the wind velocity of the ith gradation in fractions of unity.

Equations (6) establish the relation between the relative mass of aerosol fallout to the distance r_i from the BP source and the repetition of the ith gradation of wind velocity.

Estimates of w_i ($i = 1, .., 5$) in Table 2 proved to be close to each other thus confirming the feasibility of the kinematic description of the processes of PAH falling out.

Conclusion

The experimental studies performed in the vicinity of the Novosibirsk electrode-producing plant and numerical interpretation of the obtained data allow the following conclusions:

– the vicinity of the Novosibirsk electrode-producing plant is polluted with PAH mostly due to emissions from high smoke stacks of the calcining shop;

– in two-point observation plans close to optimal ones, the distribution functions of the specific PAH concentrations are determined, and their accuracy is estimated;

– the curves of the concentration distribution for most studied PAH are similar to each other, what is indicative of the composite compound of aerosol fallout and is primarily connected with peculiarities of technological processes.

The rate of the major portion of PAH falling out is estimated, and the possibility of constructing the kinematic model of PAH spreading in the vicinity of the plant is confirmed.

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