

Reconstruction of pollution fields by the receptor modeling method near Khomutovo village as an example

M.S. Zarodnyuk,¹ E.V. Kuchmenko,¹ E.V. Molozhnikova,²
I.I. Marinaite,² and O.G. Netsvetaeva^{1,2}

¹ L.A. Melent'ev Institute of Power Systems,
Siberian Branch of the Russian Academy of Sciences, Irkutsk
² Limnological Institute,
Siberian Branch of the Russian Academy of Sciences, Irkutsk

Received January 27, 2006

The problem of detecting emission sources and restoring the field of concentrations on a local scale was formalized using the data of observations (the inverse problem). The suggested statement supposes minimization of a squared discrepancy between the experimental and calculated data taking into account the balance between emissions of a pollutant over some period, its deposition on a local area, and the pollutant part transported over long distances. The observational data on chemical composition of the snow cover in industrial, agricultural, and background points of the region are used as initial ones. The observations were made at a checkpoint (Khomutovo village). The results allowed the surface, simulating the actual field of deposition, to be constructed and approximated by analytical function. It was established that for several sources of emissions the problem of calculating the coefficients is multi-extreme. The problem of searching for a global extreme of the function was solved numerically. Its real coefficients were calculated using the data on chemical analysis of the deposition (snow cover) at a reference site.

A reliable pattern of propagation of pollutants over cities or villages with a complicated relief and a developed local circulation system, for example, situated at the coast of the Lake Baikal, can be obtained only through building an original 3-D model of circulation for each such site. Additional difficulties can be brought by the absence of gradient observations at the majority of hydrometeorological stations and reliable data on the degree of atmosphere stability and the turbulent exchange intensity. Estimate of contribution of local and regional pollution sources, as well as natural and anthropogenic factors, is of great significance for the Baikal natural territory. Under these conditions, there appears an urgent necessity of solving the inverse problem: reconstruction of pollution fields for snow cover and near-ground air layer, caused by individual sources of pollution, based on data of chemical analysis of snow samples.

The way of identification of the pollution sources, applied by us, is the experimental analysis of fallouts, when the contribution of an individual source is determined using the proportion between concentrations of the prescribed set of compounds and taking into account the available data on the coordinates and power of the source (so called receptor simulation).^{1,2}

Analysis of snow cover provided for general characteristic of fallouts during the period between November and March.³ The spatial regularities revealed in the process of statistical processing can be called distribution functions. Traditionally, when constructing such functions, the normal law is used, which in the 1-D case at $x \geq 0$ is described as

$$f(x) = a \exp[-s(x - R)^2], \quad (1)$$

where a is the function maximum, R is the distance to the point of the maximal fallout, s is the coefficient inverse to the fallout intensity. The distribution functions for emissions from two different stationary sources are shown in Fig. 1.

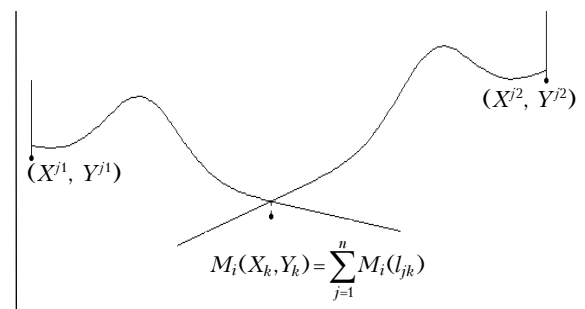


Fig. 1. Exponential distribution functions characterizing the instant effect of two sources of emission.

Actually, to determine the total fallout of i th substance at some point of a terrain, it is necessary to add the magnitudes of this substance fallout resulting from emission of each individual source.

However, when describing the fallout field for a sufficiently long period, it is necessary to take into account both the dependence of f on x (the distance from the source to the fallout receptor point) and on the angle of the source–receptor direction. If in case of a small time interval (2–30 min) the wind direction can be assumed constant, then the OX axis can be directed in parallel to wind. The pollution field maximum is located on this axis at the distance R . A decrease of concentration of some pollutant (or fallout amount)

in the direction perpendicular to OX is also described by the normal law. Obviously, the wind direction cannot be constant during the period of several months.

Note the following properties, characteristic of the actual field of the fallout from some individual source of emission. First, the point, corresponding to the coordinates of the source, is stationary and corresponds to the distribution function minimum equal to zero. Second, if the distance between the source and the receptor tends to infinity, the function values also tend to zero. Third, according to observational data, the function has two local maxima in the most repeated, almost adjacent directions from the source. At last, it should correspond to the normal distribution law at some part of the superposition, which does not contradict to the second and third properties.

Taking into account the above peculiarities, the following function is suggested as the distribution of i th component from j th source:

$$F_i(X, Y) = [A_i((X^j - X)\cos\varphi - (Y^j - Y)\sin\varphi)^2 + B_i((X^j - X)\sin\varphi + (Y^j - Y)\cos\varphi)^2] \times \exp[-s_i(\sqrt{(X^j - X_i)^2 + (Y^j - Y_i)^2} - R_i)^2], \quad (2)$$

where A_i/B_i is the coefficient of the direction prevalence, (X_i, Y_i) are the coordinates of their deviations from adjacency, φ is the wind direction angle. Figure 2 shows the distribution function surface of the pollutant fallout from one source, characteristic of the south of Eastern Siberia. The distribution of one component from all sources is represented by the sum of functions (2) for all $j = \overline{1, n}$.

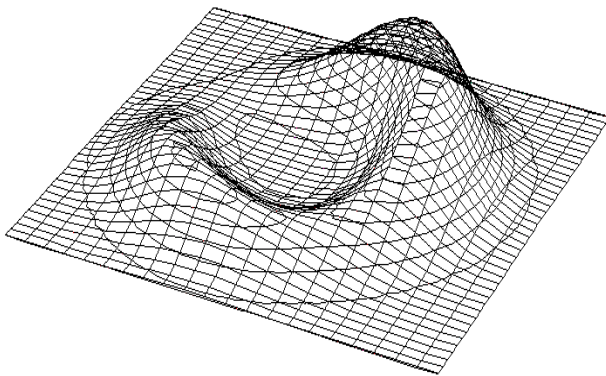


Fig. 2. Surface of the pollutant distribution function in snow cover characteristic of the south of Eastern Siberia.

There is a linear dependence between the substance emitted during some period, its local fallout, and the fraction caught by the remote transfer.² From the mathematical standpoint, such relationships set a system of linear equations:

$$M_i(X_k, Y_k) = \sum_{i=1}^m Q_{ij}x_{jk} + \sum_{i=1}^m G_i - \sum_{i=1}^m Q_{ij}P_j, \quad (3)$$

where $M_i(X_k, Y_k)$ is the mass of i th pollutant at k th measurement point, Q_{ij} is the emission of i th pollutant by j th source, x_{ik} is the contribution of this source to k th measurement point, $k = \overline{1, r}$; P_j is the fraction of j th source emission carried over by the remote transfer out of the populated territory, G_i is the contribution of the atmosphere background pollution into the fallout at the receptor point for i th pollutant.

To find parameters of the distribution function from the observation data, it is necessary to solve the following problem: to determine

$$\sum_{k=1}^r \left(\sum_{i=1}^m M_i(X_k, Y_k) - m \sum_{j=1}^n F(\alpha_j, X_k, Y_k) \right)^2 \rightarrow \min_{\alpha} \quad (4)$$

at the restrictions

$$a_{ij} > 0, s_{ij} > 0 \text{ and } R_{ij} > 0, \\ 0 < R_{ij} < X^j - Y_i, 0 < R_{ij} < Y^j - Y_i, \quad (5)$$

where $\alpha_j = (A_j, B_j, \varphi, s_j, X_j, Y_j, R_j)$.

In order to decrease the dimensionality of the problem, it is necessary to study the dependence of the function parameters on technological characteristics, such as the source height, temperature and intensity of emission, as well as meteorological and geographical parameters, which in some cases can be assumed identical for all sources on the populated area. Then the relation (4) can be applied to a great number of sources almost without the loss of accuracy.

Generally speaking, determination of distribution function parameters is not a key stage in the framework of statement of the problem of identification of the pollution sources. There is a possibility of simultaneous calculation of the distribution function parameters and the effect of the sources on the pollutant content at the receptor points. Based on the assumption about equal spread of light-weight pollutants, we have

$$\sum_{k=1}^r \left[\sum_{j=1}^n \left(\sum_{i=1}^m Q_{ij}x_{jk} - \sum_{i=1}^m Q_{ij}P_j + \sum_{i=1}^m G_i - mF(\alpha_j, X_k, Y_k) \right) \right]^2 \rightarrow \min_{\alpha, x} \quad (6)$$

accounting for restrictions (3)–(5).

On the one hand, the statements (3), (5), and (6) can be classified as the problem of approximation based on the least squares method. Therefore, the number of approximation points with accounting for the restriction equalities should be greater than the number of variables. On the other hand, the proposed assumption can be considered as the problem of global approximation, which solving by numerical methods requires studying of the solution uniqueness.⁴

To improve parameters of the distribution function, the sampling was carried out at a reference site as well. When selecting the reference site (Khomutovo village), we took into account the number of

pollution sources, peculiarities of the relief, and the local circulation. The village is situated 30 km to the north-northwest from the center of Irkutsk and far away from large water objects (Fig. 3). Then we assumed that admixtures from local sources in Khomutovo are spreading over a flat homogeneous surface, and can be described by standard techniques with a satisfactory accuracy. According to ground-based and aerological data, repetitiveness of southern winds is quite low (especially in the cold season), therefore the direct effect of Irkutsk on the village can be ignored.

The main sources of atmospheric pollution in the village are two boiler-houses, using the Azeisk brown coal. Their total power is 3 Gcal/hour. To heat private houses (brick stoves), firewood and coal are used.

The snow sampling was carried out in Khomutovo on February 21–22, 2005. A total of 32 snow samples were collected. The sampling map is shown in Fig. 3. Collection of samples for analysis of ions and the content of polycyclic aromatic hydrocarbons (PAH) was carried out in parallel. The relative content of

PAH in the samples normalized to benz(a)pyrene is presented in Table 1.

As is seen in Table 1 and Fig. 4, the composition and the ratio of individual PAHs in fall-outs in Khomutovo are, on the average, close to the background and essentially lower than, for example, in Slyudyanka, where more than 20 heat sources are situated.

Nevertheless, like in Slyudyanka, the increasing content of fluoranthene and phenanthrene is noticeable, that is characteristic of snow cover in zones of action of small boiler-houses. Fluoranthene and phenanthrene significantly exceed the background values at points No. 3 (gate) and No. 8 (school) situated at the distance of 40–50 m from the chimney of the central boiler-house (No. 3) and approximately 20 m far from the school boiler-house (No. 8). Thus, the study of concentrations of individual PAHs even by the expert estimate method allows us to confidently identify the effect of individual local heat sources against the background of the regional pollution by big industrial centers.

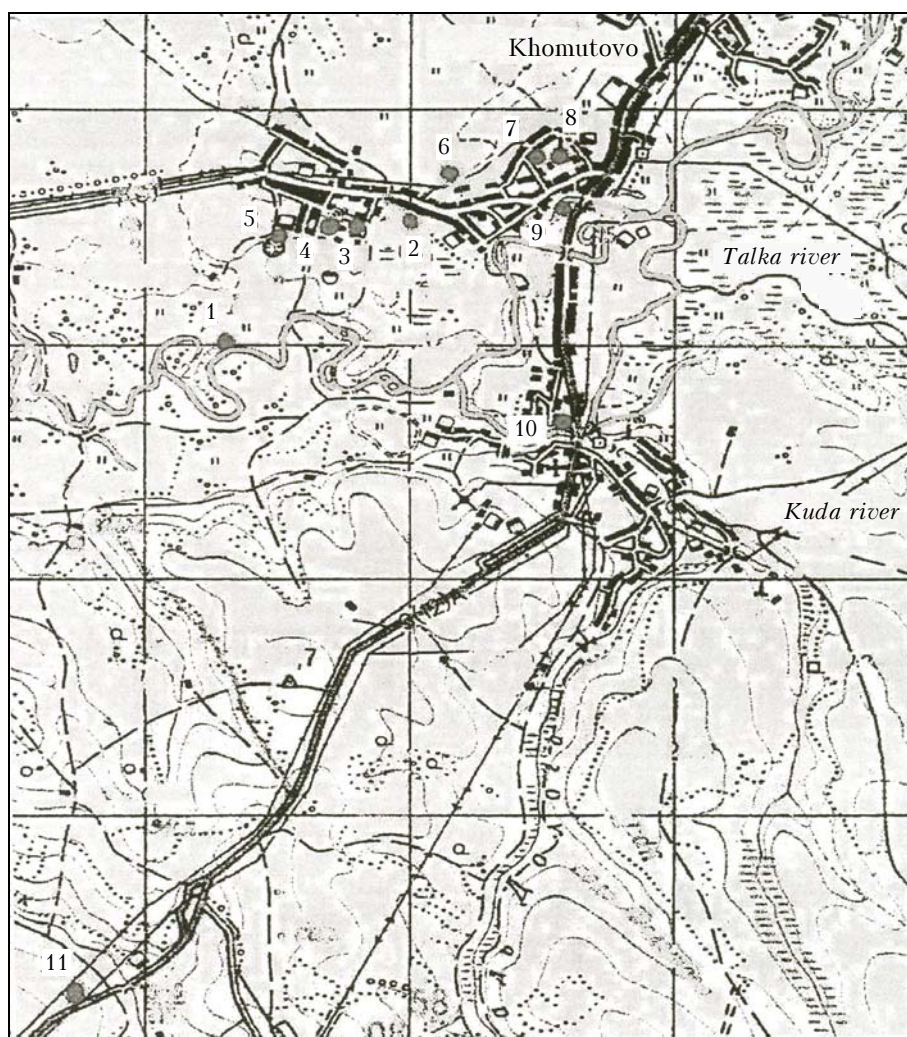


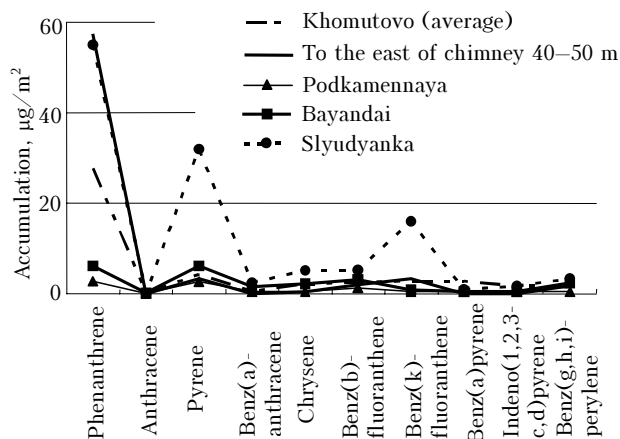
Fig. 3. Map of sampling in Khomutovo.

Table 1. Fraction of PAH in fall-out in Khomutovo

No. of sample	Place of collection	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Benz(a)-anthracene	Cryzehe	Benz(b)-fluoranthene	Benz(k)-fluoranthene	Benz(g,h,i)-perylene	Indenopyrene
1	Outskirts	14.24	0.09	5.72	2.84	0.00	1.73	2.45	1.60	0.00	0.00
2	Beyond wetland	11.26	0.12	2.64	1.67	0.00	0.00	1.36	2.57	5.17	2.99
3	Gate	32.15	0.00	3.88	1.89	0.00	0.24	1.10	1.90	0.00	0.00
4	Dump	17.96	0.00	3.61	1.34	0.00	0.00	0.00	0.00	0.00	0.00
6	West	13.77	0.00	6.94	3.91	1.64	2.05	1.64	2.34	2.21	1.70
8	North	30.55	0.54	10.52	6.68	3.03	3.84	4.03	4.51	4.82	3.29
9	Northeast	6.63	0.08	3.31	2.10	0.48	1.34	1.63	1.58	1.76	1.07
10	School	9.62	0.15	3.37	1.85	0.29	2.19	1.40	1.71	3.91	2.83
11	1st bridge	11.98	0.18	5.68	3.04	0.37	2.61	1.85	1.59	2.13	1.33

Table 2. Fall-out of main ions calculated from the results of chemical analysis of samples collected in Khomutovo, mg/m²

Place of collection	HCO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻	Cl ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	NH ₄ ⁺
Outskirts	551.0	332.0	79.1	315.9	178.3	11.0	213.5	27.0	37.0
Beyond wetland	194.8	125.1	43.3	14.8	7.2	2.9	77.9	11.5	14.8
Gate	1379.7	425.3	134.0	30.2	25.7	32.1	469.8	47.0	30.2
Dump	895.2	422.9	106.7	11.0	17.1	9.9	354.3	30.5	22.9
West	2300.0	582.7	176.3	26.1	21.5	98.1	682.3	75.1	62.9
North	142.5	125.1	65.2	30.1	9.2	5.1	80.4	13.6	19.6
Northeast	38.4	112.5	61.3	10.6	3.4	4.7	50.6	8.8	18.1
School	51.3	160.7	76.1	45.2	23.7	6.9	63.1	10.7	30.6
1st bridge	139.3	190.7	64.2	40.7	25.7	8.8	78.8	15.4	23.8
3rd bridge	72.9	258.7	140.5	246.3	146.4	14.1	104.1	17.0	44.7
Road to Irkutsk	0.32	30.0	151.7	59.2	231.0	127.2	13.1	58.9	13.1

**Fig. 4.** Composition of PAH in snow cover of different sites situated in the Angara river region.

The results of ion analysis of samples collected in Khomutovo are shown in Table 2.

As is seen, the enhanced pollution by sulfates is observed only in zones of acting of the central boiler-house in directions to the east (point No. 3) and to the west-west-south (point No. 4).

The factor analysis carried out using the standard software package STATISTICA 6.0 allows us to select four factors: HCO₃⁻, SO₄²⁻, NO₃⁻, K⁺, Ca²⁺, NH₄⁺, Mg²⁺ ions, evidently, characteristic of the boiler-house emissions, as well as fluoranthene

and phenanthrene; Na⁺ and Cl⁻, most probably, having a natural origin; all other PAHs, except for benz(a)anthracene, benz(g,h,i)perylene, and indenopyrene are characteristic of the regional background taking into account the fact that all samples were collected within 100 m from highways. The benz(a)anthracene, benz(g,h,i)perylene, and indenopyrene are separated as an individual factor, perhaps, because their concentrations in samples are very low and, hence, the obtained characteristics are insignificant. The sulfates data in fallouts at the reference site allowed us to determine the parameters of the real distribution function for fallouts from an individual source of emission on the local scale (Table 3).

Table 3. Parameters of the function (2) for the central boiler-house, obtained numerically using actual data on the sulfate fall-outs

Parameter	Scale	
	global	local
<i>A</i>	11485.26	12773.27
<i>B</i>	503.55	535.26
<i>φ</i>	0.5	3.63
<i>s_j</i>	0.64	0.61
<i>X_j</i>	415.46	415.52
<i>Y_j</i>	756.66	756.62
<i>R</i>	0.12	0.102

The obtained results can be useful in solving the problem of identification of contributions of the emission sources.

Acknowledgments

This work was supported in part by the Russian Foundation for Basic Research (grant No. 05–05–97233r-Baikal-a).

References

1. E.V. Kuchmenko, E.V. Molozhnikova, and S.P. Filippov, *Atmos. Oceanic Opt.* **15**, Nos. 5–6, 490–493 (2002).
2. E.V. Kuchmenko, E.V. Molozhnikova, I.I. Marinaite, and M.S. Zarodnyuk, *Atmos. Oceanic Opt.* **18**, Nos. 5–6, 446–449 (2005).
3. V.F. Raputa and V.V. Kokovkin, *Khimiya v Interesakh Ustoichivogo Razvitiya*, No. 10, 669–682 (2002)
4. O.V. Vasil'ev, *Lectures on the Methods of Optimization* (State University, Irkutsk, 1994), 344 pp.