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Elaboration and Testing of a Local Model of Deposition of Industrial Pollutants from the Atmosphere onto the Ground Surface

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Abstract

On the basis of the known theoretical developments, a sufficiently simple model of local scale has been built and software-supported, which uses as the input information the normative databases on industrial sources and accessible climatic information. Bibliographic sources and general principles of model construction are considered, and the calculation results are compared with data of experimental studies of some industrial objects and cities of the Kemerovo Region.

INTRODUCTION

When substantiating the system of ecological monitoring in [1], it is underlined that it is based on observations which (as far as the atmosphere is concerned) include both the atmospheric pollution (AP) and the main cause of pollution – emissions of pollutants of industrial sources into the atmosphere. Observation of AP is carried out by a rather sparse network of stationary posts in large industrial cities on an insignificant part of pollutants (5–20 ingredients). Observations of industrial emissions into the atmosphere are represented by the inventory data. The inventory is implemented mainly using balance calculations uniform for all branches of industry. The calculations are carried out under the control of environmental authorities.

Besides, the most important components of monitoring are calculations of atmosphere pollution by the emission parameters. Indeed, it is possible to forecast the AP level for planning the industrial development only by means of calculation methods. In the general case, instrumental observations of pollutant concentrations cannot estimate the contribution of a concrete atmosphere pollution source (APS) to the total measured amount, which is necessary for establishing the standards for permissible emission rates (PER) and counting the emission fee. That is why modeling is necessary both for perspective and for the current period. There are various models [2, 3], but (for the sake of uniformity and comparability), for regulatory applications (designing works and PER estimation), in Russia a unified model [4] for calculation of maximal short-term (averaged for a 20-minute interval) ground-level concentrations arising under the conditions of atmospheric diffusion (unfavorable but rather frequent) and characteristic of the given territory has been established. The model is built on the basis of a numerical solution of a stationary semiempirical turbulent diffusion equation with subsequent analytical approximation of the results for various types of sources. The programs that realize [4] for computers are approved for regulatory calculations only after a special testing and coordination with Nature Resources Ministry at the federal level [5].

A considerable part of all pollutants is made up of particulate matter having a sedimentation effect on the ground surface. Lately, when carrying out ecological expertise of some projects, problems of differential and integral estimation of the amount of emitted pollutants (EP) falling out from the atmosphere onto the given territory during rather long time periods (a season, a year *etc.*) have been raised. For example, in the Kemerovo Region, such problems arise when estimating the wash-out of atmospheric pollutants in the reservoir being designed on the Tom' river, the deposition of industrial pollutants of coal mining industry onto agricultural lands *etc*.

In the present work, a model for calculation of EP deposition and attempts to apply it for solution of practical tasks are presented. The model has been implemented as an additional utility of a software complex ERA which has been approved as corresponding to the model [4] and fit for using it in regulatory applications (for more details, see www.logos-plus.ru). This permits using the standardized databases accumulated in ERA, a finished-off user interface, and all the textual and graphical capacities of the complex on presenting the output information (including that covering on digital maps and raster substructure).

MODEL OF DEPOSITION OF CONTAMINANTS ON THE GROUND SURFACE

General principles of model construction

For quantitative estimation of deposition of particulate matter emitted out of industrial sources onto a given territory, a model based on the procedure [6] issued by the Institute «Atmosphere» (Ministry of Natural Resources of Russia, St. Petersburg) has been developed and realized in software-supported manner. The procedure is based on works [7–9] and permits estimating the mean annual ground-level concentration, the wet flow of EP on the ground surface P_w (removal), dry flow (deposition) P_d and the full flow P which represents the sum of the two mentioned above components ($P = P_\mathrm{w} + P_\mathrm{d}$), by the data on the parameters of the sources, on the fraction composition of the emitted pollutants and on the climatic meteorological parameters of the territory.

Calculation of the mean annual wet flow P_w, g/m^2 per year, of pollutants emitted by a single point APS onto the ground surface at the given point is carried out by the formula:

$$P_\mathrm{w} = \frac{(1+b)M}{2\pi u r L_0}\left[aL_\mathrm{s}t_\mathrm{ws}\sum_{i=1}^{K} m_i y_i \exp\left(-\frac{a y_i r}{\bar{u}}\right) + L_\mathrm{w}t_\mathrm{ww}\sum_{i=1}^{K} m_i y_i \exp\left(-\frac{y_i r}{\bar{u}}\right)\right] \quad (1)$$

where b is the relative contribution of mixed atmospheric precipitation to their total amount; M is the mass of particulate matter emitted by the source into the atmosphere during a year, g/year; $\bar{u}$ is the mean annual wind speed in the diffusion layer, m/s; L_0 is the recurrence of the given wind direction for the circular wind rose; a is the empirical correction factor for the difference in the intensity of removal by liquid (rain) and solid (snow) precipitation; L_s, L_w are the recurrence of the wind direction of the given point for the summer and the winter wind rose, respectively; t_ws, t_ww are the relative (in parts of the year) period of liquid and solid precipitation; K is the number of particle fractions considered; m_i is the proportion of the total pollutant mass per the i-th particle fraction; y_i is the constant of wet removal of the i-th particle fraction, s^{-1}.

The mean annual dry flow P_d, g/m^2 per year, is estimated as the sum

$$P_\mathrm{d} = \sum_{i=1}^{K}(V_{i\mathrm{w}}t_\mathrm{dw} + V_\mathrm{is}t_\mathrm{ds})q_i \quad (2)$$

where $V_{i\mathrm{w}}$ is the rate of sedimentation of the i-th particle fraction in winter, m/s; t_dw is the period of snow cover minus the time of precipitation at this period, s; $V_{i\mathrm{f}}$ is the rate of sedimentation of the i-th particle fraction onto the surface free from snow cover, m/s; t_ds is the duration of snow cover-free period minus the time of precipitation, s; q_i is the mean annual ground-level concentration of the i-th fraction, g/m^3.

For calculation of the field of the average annual concentration q_i (function of r, M, m_i, geometrical parameters of APS, temperature and emission rate, climatic parameters), the approach and approximation formulae presented in [6] were used, according to which the average annual concentration is re-calculated by the maximal short-term one estimated on the basis of the regulatory method [4]. For recalculation, such parameters as the average annual wind speed $\bar{u}$, the wind rose and the repeatability of the conditions determining the vertical and the horizontal dispersion in the atmospheric boundary layer (estimated in [4] by the coefficient A) are used. The calculation of the average annual concentration is made separately at each receptor point of interest, for each fraction of particles with various sedimentation parameters, whereupon the results are added to obtain the total concentration for each APS.

The total flow of pollutants emitted by the set of APS is defined at each receptor point as a superposition of contributions of separate sources which practically may be point, linear or area. For linear and area APS, integration with a source length or area step depending on the distance r is used. The step size is chosen on condition of not exceeding the calculated error of 3 %. One has to underline that the software-supported realization of the model under consideration envisages using individual meteorological parameters and wind rose for each APS, depending on the belonging of the source to the concrete city.

Relations (1) and (2) show that within the framework of the assumed model one can easily estimate the seasonal deposition. In particular, for calculation of pollutants deposition into the snow cover (winter period) it is sufficient to assume $t_{ws} = t_{ds} = 0$ and to use the total emission rate M for each source separately only for the winter period. When calculating the average winter concentrations q_i, one has, naturally, to use only the winter wind rose L_w and the average winter wind speed.

Subdivision of settled contaminants according to fraction composition

In [6, 7] subdivision of industrial discharges into 12 fractions according to particle size is considered. For the first numerical experiments considered further on, 5 fractions which parameters are presented in Tables 1 and 2 have been left.

TABLE 1

Subdivision of discharges of an industrial city according to fraction composition

Fraction code	Size class	Aerodynamic diameter, μm	Content in fractions, %			
			aitken	aggregations	course	total
8801	Very small	<1	100	96.4	1.4	41.2
8802	Small	1–10	0	3.6	42.2	25.8
8803	Middle	10–50	0	0	46.1	27
8804	Large	50–100	0	0	7.3	4.3
8805	Very large	>100	0	0	3	1.7

TABLE 2

Parameters of elimination of particles from the atmosphere onto the subjacent surface

Fraction code	Constant of wet removal, $y_i \cdot 10^{-4}$, s^{-1}	Rate of deposition onto the snow, V_{iw}, m/s	Rate of deposition onto the soil, V_{is}, m/s	Parameter F according to OND-66	Size class
8801	0.01	0.001	0.010	1.0	Very small
8802	0.70	0.007	0.013	1.1	Small
8803	3.83	0.042	0.043	1.5	Middle
8804	4.48	0.151	0.155	3.0	Large
8805	5.00	0.420	0.430	4.8	Very large

With the exception of small distance from the source, the main contribution is made by dry deposition [8] which depends on the average annual concentration. Within the framework of the assumed model of calculation of average annual concentrations, the dependence on sedimentation processes is described only by the parameter F which, (according to [7]), for particles of 0.1 to 20 μm is equal to 1.0, and for those of 20 to 50 μm is equal to 1.5. As a result, 5 fractions differing in parameter F remain. Besides, in practice, the fraction composition of emitted pollutants cannot be estimated not only as exactly, but is not measurable at all, and therefore information about it is not available in standard databases for industrial APS.

Without special additional studies, one can obtain only approximated expert estimates of the fraction composition for various source types, proceeding from the type of industrial process and depending on the availability in the APS of particulate control equipment which separate mainly large and middle-size particles. That is why for the first experiment with carrying out practical calculations, subdivision into fractions was performed using a model of urban aerosol distribution across fractions unified for all APS [10], assuming that the aggregated area source (city) would have such a distribution after mixing the emissions of all the internal individual sources. Modeling is made for three regions of particle sizes with different mechanisms of origin: Aitken or generation fraction, accumulation fraction, coarse fraction.

In Table 1, the particle content calculated according to the class size, and their total content are presented. The total content determines the subsequently used fraction composition of settling pollutant for APS emissions of an industrial city. Therein, it is assumed that the volume contribution of three particle types to the total amount of city aerosol is equal to 11.25, 30.15 and 58.6 %, respectively [11]. The density of particles of all the fractions is assumed to be the same and equal to 2.16 g/cm^3 [11]. The rates of sedimentation onto the snow and onto the soil can be calculated by the formulae presented in [7].

For implementation of calculations taking into account the fraction composition, five pollutants with codes 8801–8805, sedimentation coefficients according to Table 2, and MPC equal to 1 mg/m^3 were added to the pollutant database of the software complex ERA. The latter equation permits using all the calculation blocks of ERA complex for obtaining the average annual concentration in milligrams per cubic meter (for regulatory calculations, the results are expressed in the parts of the Air Quality Standard for each pollutant) and therefore the flow of settling particles in grams per square meter per year (season). After setting new pollutants, an imaginary summation group* of the indicated substances was formed, which makes it possible to use the method of differential calculation of the total concentration realized in the program ERA and applicable for summation groups of pollutants with different sedimentation coefficients F according to [4]. Thereby, the requirement of [6] concerning calculation of settling by separate fractions with subsequent summation is fulfilled automatically.

The parameters $V_{i\mathrm{w}}$ and V_{is} presented in Table 2 have the dimension of velocity, depend on the size and density of particles and on the roughness of the ground surface (snow is considered as a surface with low roughness, and soil as having a moderate one). The sedimentation rates have been obtained on the basis of empirical formulae and presented in [7] as tables for 12 fractions with more detailed subdivision of particles with respect to size. In the data of Table 2, the 12 fractions from [7] are united into 5 fractions used for subsequent calculations, with averaging the parameters $V_{i\mathrm{w}}$, V_{is} and y_i. It is noteworthy that the independence of sedimentation rates of spatial coordinates presupposes uniformity of the ground surface, which may become a cause of error in modeling deposition in a concrete point.

Possibilities of the model

All the above mentioned calculated schemes have been realized in software-supported way

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*According to Russian Sanitary Law some pollutants with unidirectional harmful influence form summation groups that must be combined as a single pollutant for air quality estimates.

in the ERA software complex. This permits carrying out practical calculations using standardized databases both for separate industrial objects and for any combinations thereof (city, region *etc.*) up to a task scale of 100 km. Calculation zones are preset on the plane in Cartesian coordinates and may represent a rectangle, an arbitrarily outlined (singly or multiply connected) region, a line (or a set of lines) of arbitrary form, or a set of fixed points. All the zones may be set graphically on a digital map of the territory. The rectangle, region or line are automatically covered with a net of a preset regular step. Besides, all the inflection points for the line and the region borders are considered as calculated. The number of calculated points and sources is practically unlimited and is determined by the computer parameters.

For calculation of pollutants settling onto the surface, the function of automated subdivision of summed amount of particulate mass emitted from industrial sources according to their fraction composition depending on the number of fractions preset by the user and the contribution of the given fraction to the total emission (see Table 1) is incorporated into the software. This permits getting rid of a large number of data handling and additional calculations when solving practical tasks where the number of sources is counted by the hundred. There may be any number of fractions, but in the examples shown herein below it is 5. The program previews a linear smoothing of the 8-point wind rose.

ESTIMATION OF APPLICABILITY OF THE MODEL TO REAL PROBLEMS

A natural stage in estimation of applicability of the model is a comparison of the modeling results with observation data. The most accessible are the results of analysis of snow samples with respect to the content of separate pollutants or the sum of suspended particles. Therein, it is necessary to have information about APS parameters and about emission rates of controlled pollutants from those industrial objects which influence zones include the control points. Below, three examples are considered for the Kemerovo Region for which both information about the emission rates and data of snow analysis are available.

Zinc salts' content in the snow in the vicinities of Belovo

In [12], data of analysis of the snow cover carried out by the Institute of Soil Science and Agrochemistry, SB RAS, in 1987–1988 for the purpose of estimating the amount of zinc accumulated in snow in the vicinities of the Belovo Zinc Plant are presented. APS parameters and discharge values for the zinc plant were borrowed from a summarized project of PER standards for Belovo (Kemerovo Region) which corresponded to 1999. It is noteworthy that in APS inventory data for Belovo, zinc salts' emissions from other enterprises are not available. This means that the ratio of enterprises“ emission to Air Quality Standard of zinc salts is no more than 0.05. Since this ratio for the sum emission of APS of the zinc plant is more than 10, the emissions of the rest of sources make up less than 1 % and do not influence considerably the contamination of the territory under study. The types, altitude and location of APS have not undergone any radical changes during this time. The authors have no data on the total production by the plant in 1988 and 1999; nevertheless, one may say that all the sources of the technological line that has not changed make their contributions proportionally to the power of the plant. That is why one may expect that comparison of measurements and calculations will result in obtaining something like curves of dependence of precipitation on the distance from the plant, independently of the annual production of the enterprise.

For calculations, the winter wind rose for the observation period [12] was taken, and unified values for climatic parameters of the sedimentation model were assumed: $\bar{u}$ = 4.1 m/s; t_{ws} = 0; t_{ww} =0.17; t_{dw} =156; t_{ds} = 0; a = 1; b = 1, which corresponded only to the winter period for Belovo [13]. The values of corrections a and b correspond to the case of absence of any differences in wet removal by solid and iquid precipitation. The general picture in

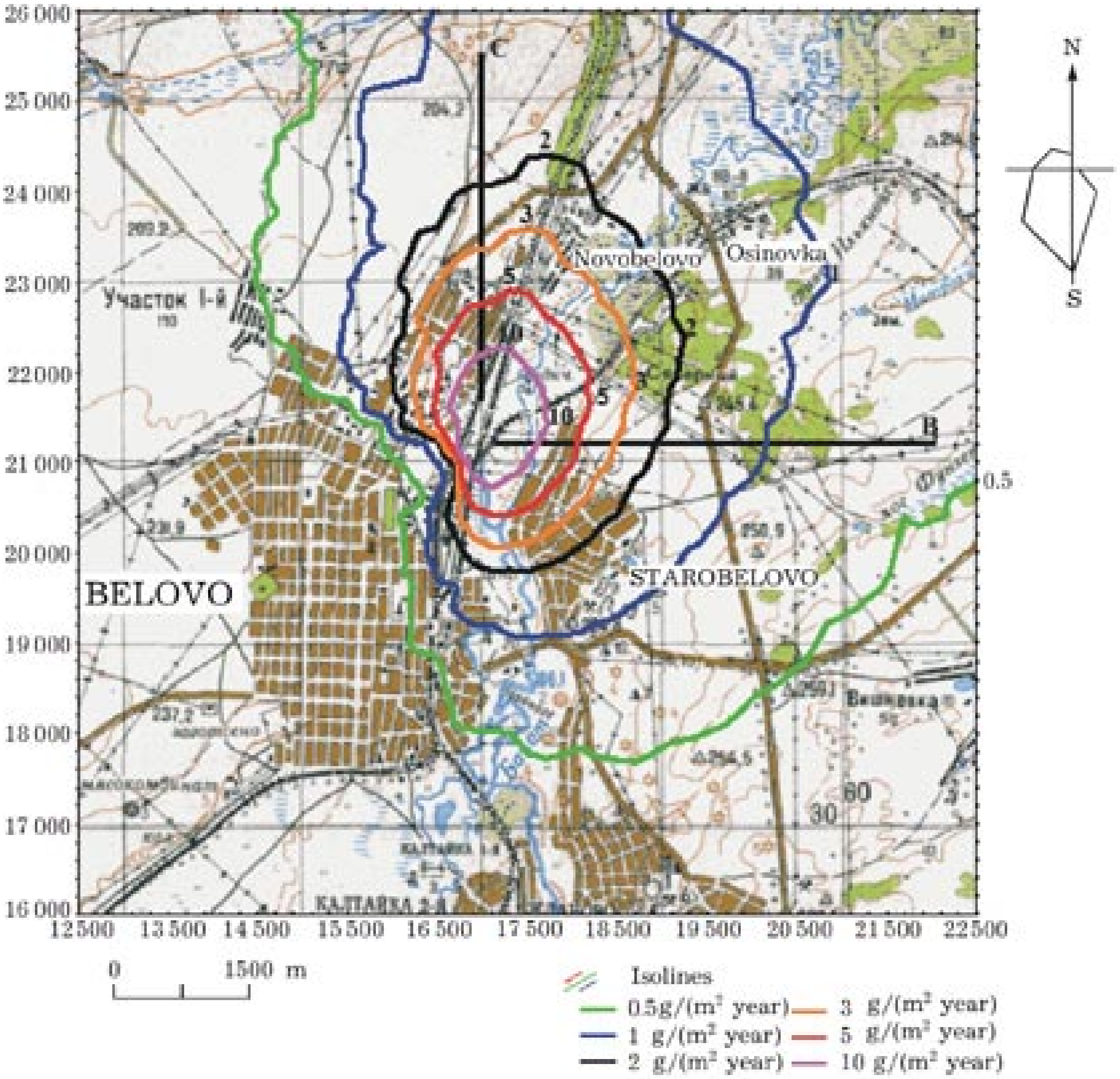

Fig. 1. Calculated estimate of zinc salts' accumulation in the snow in the vicinities of Belovo (a raster map).

the form of deposition isolines for zinc salts constructed by the results of calculation on a rectangle with the size of 10 × 10 km with a net step of 200 m is shown in Fig. 1.

Here, two directions (northwards and eastwards) are distinguished for which a comparison of calculated values for the winter period with the data of analysis of the snow cover borrowed from [12] has been carried out. The comparison results are presented in Table 3 and in Fig. 2.

The measurements for each of 8 wind direction points were performed at 7 points distant from the sources at 0.5–10 km. The observation data are presented in terms of background concentration which absolute value is not given. This is the second reason for which there is no sense to compare the absolute values presented in Table 3 (the first reason is the temporal non-coincidence of data on emissions with on-location observations).

The results of comparison of calculated and experimental data presented in Table 3 and Fig. 2 show that the patterns of changes of zinc salts deposition depending on the distance from the sources coincide rather well, with the exception of close distances (below 1 km), where the model gives a noticeable relative excess over the data of on-location observations.

TABLE 3

Sedimentation of zinc salts in the vicinities of Belovo (measurement and calculation)

Point No.	Distance, km	Northward direction		Eastward direction	
		Measurement*	Calculation, g/m^2	Measurement*	Calculation, g/m^2
1	0.5	281.00	34.95	68.40	9.82
2	1.0	155.00	13.08	49.80	5.03
3	2.0	45.00	4.02	24.80	1.76
4	3.0	16.00	2.07	18.40	1.04
5	5.0	13.00	1.04	1.90	0.56
6	8.0	–	0.60	5.40	0.33
7	10.0	5.80	0.47	4.20	0.26

*Deviation from the background.

Suspended substances' content in the snow in the vicinities of Bekovo settlement

For estimation of the influence of industrial cities and coal mining enterprises of the Kemerovo Region on the pollution of agricultural lands, experimental and calculated estimations of the total dust deposition for the winter period of 2001 were carried out in the vicinities of the settlement Bekovo. Analysis of snow was carried out at 5 control points. Calculated estimates were obtained on the basis of standard data of emission into the atmosphere and of the values for climatic parameters for the winter period in the vicinities of Belovo.

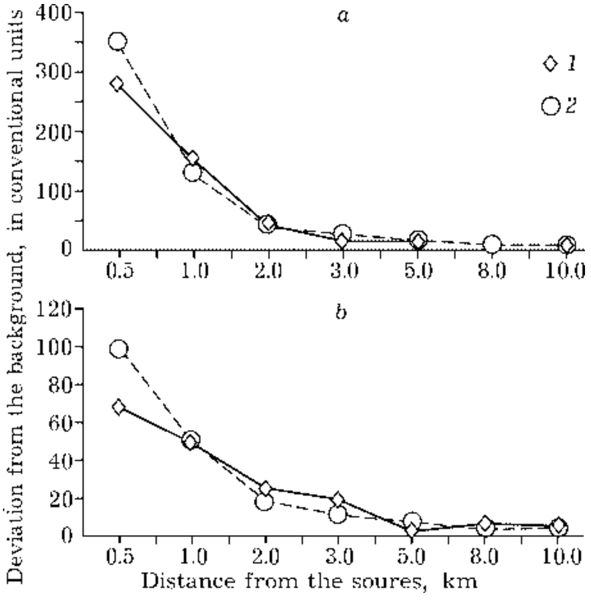

Fig. 2. Change of measured and calculated characteristics of zinc salts deposition depending on the distance from the sources in the northward (*a*) and eastward (*b*) directions: *1* – measurement, *2* – calculation (×10).

When making calculations, the cities of the Kemerovo Region situated far from the settlement Bekovo are represented by aggregated area sources with the total emission of particulate mass. Plausibility of such a comparison for distances of more than 30–40 km from the city has been demonstrated in [14]. Calculation made only for remote cities permits concluding that the flow of dust particles onto the territory under study is practically constant and amounts to approximately 2.7 g/m^2 for the winter period. This value may be assumed as a calculated estimate of the constant background deposition (Table 4) which one has to add to the results of calculation of pollution from nearby Industrial objects (Belovo and Guryevsk cities in the coal mining complex of the Bachatsk pit). Results of detailed calculations taking into account the concrete location and the type of APS of nearby industrial objects and the data of snow analysis at 5 control points are presented in Table 4. Their comparison in the initial form and after multiplication by the empirical multiplier 0.215 is shown in Fig. 3.

One can see that the calculated estimates exceed considerably (by about 5 times) the results of analysis. An exception is only point 1 which is windwards with respect to the complex of enterprises of the Bachatsk pit (the repeatability of the wind direction from the sources to the point in the wind rose is minimal). The isolated location of this point in the graph can most probably be accounted for by the presence of not taken into account in the

TABLE 4

Results of analysis of snow samples and model estimates of deposition of suspended substances at control points for the winter period

Parameter	Number of the control point				
	1	2	3	4	5
Content of dust in the snow sample, mg/l	14.00	19.60	21.00	26.00	18.40
Experimental estimate of deposition onto the snow for the winter, g/m^2	4.10	5.70	6.10	7.60	5.40
Calculated estimate of deposition onto the snow for the winter, g/m^2	3.98	23.62	26.79	31.81	23.01
Calculated estimate of the background for the winter, g/m^2	2.7	2.7	2.7	2.7	2.7
Sum of calculated values for the winter, g/m^2	6.68	26.32	29.49	34.51	25.71
Experiment/calculation result ratio	0.595	0.217	0.202	0.220	0.219

calculation but really existing sources of particulate matter. Nevertheless, the total (with the exception of point 1) course of measured and calculated values coincides rather well. This gives a certain assurance that the main factors and the initial values determining the processes of dust deposition onto the surface are detected by the model plausibly and after an appropriate (see Fig. 3) calibration the model may become applicable both for estimation of the total fallout of dust emissions onto the surface and for finding the main factors and culprits of pollution. The causes of considerable discrepancy between the calculated and measured values may be faults of the model, inaccuracy of the initial data (on the sources, meteorological parameters, fraction composition etc.) or measurement errors. Most probably, all the mentioned causes are present simultaneously. To answer more concretely, a detailed analysis both of data of on-location observations and of the peculiarities of constructing the deposition model, as well as the description of interaction with the ground surface are necessary.

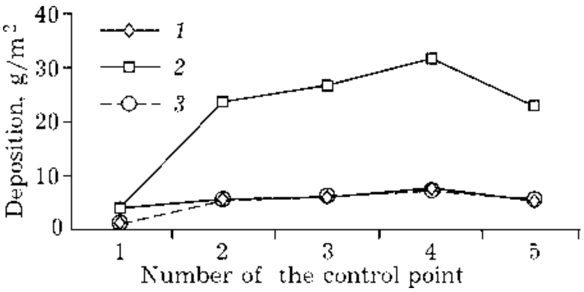

Fig. 3. Comparison of calculated fallout of dust pollutants for the winter in the settlement Bekovo with the data of snow analysis at control points prior to and after the calibration: *1* − measurement, *2* − calculation (×0.215).

Estimation of dust deposition onto the snow for Kemerovo

The dispersed composition of atmospheric pollution deposition onto the snow cover was studied for Kemerovo in winter 1994–1995 according to the research plan of the Kemerovo Science Center, SB RAS (M. Yu. Klimovich). Samples were collected in late winter before the beginning of the spring snow thawing on an area of 10 × 10 m^2 by separate core samples with the condition of obtaining a weight of dust particles of no less than 1 g. When taking samples, the researchers were guided by the RD 52.04.186–89, Pt. 2, Section 5 “Observations on the Pollution of the Snow Cover”. The dispersed composition was studied using a set of sieves with the meshes of 600, 250, 150, 100, 63, and 44 mm. Fraction of 44–2 mm was obtained by sedimentation, and fraction of 2–0.2 mm by centrifugation. Fraction weights were dried in a desiccator at 103 °C and kept under room conditions for 24 h before being weighed on scales VLR-200. The temperature and humidity of the room conditions were controlled and were found to be stable. The total dust loads in the sites of sampling in

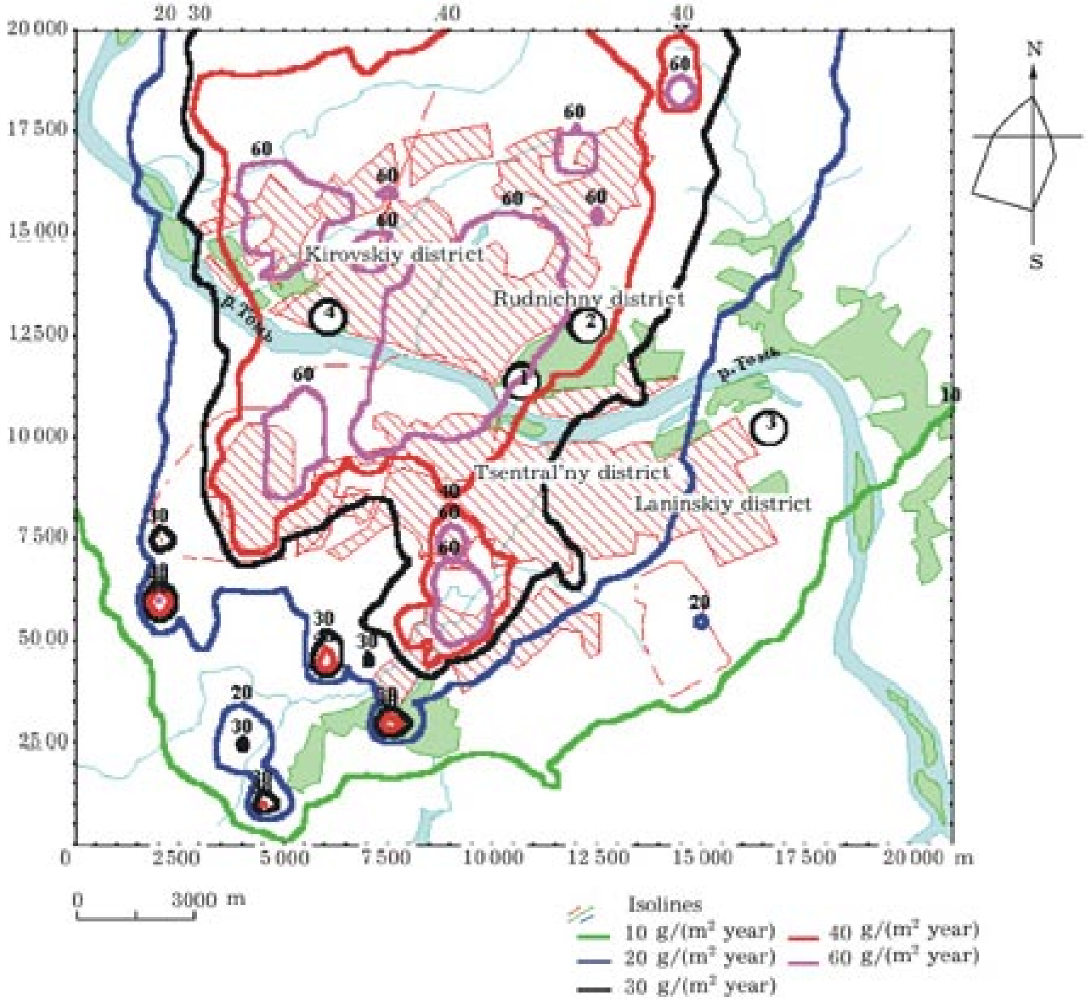

Fig. 4. Digital map of Kemerovo with isolines of calculated precipitation of solid particles onto the earth surface. In black circles, points of snow sampling are numbered.

various districts of Kemerovo amounted to 87, 108, 23, and 129 kg/(km^2 day). In this way, according to the experiments, during the time between the stabilization of the snow cover and the beginning of the spring thawing, 11.7, 15.5, 3.1 and 16.3 t of dust particles per 1 km^2, respectively, fall out from the atmosphere in these sites.

In 1997, a joined city design of PER for stationary atmosphere pollution sources of Kemerovo was compiled. Standardized databases for pollution sources refer to 1996 and are compatible with the software complex ERA. On the basis of these data and of climatic parameters of Kemerovo [15], calculations of dust deposition from the set of all the industrial enterprises included in the inventory (about 1500 sources) were carried out for a calculated rectangle of 21 x 20 km with a step of 500 m (Fig. 4) and for 4 sites where snow analysis was carried out.

In Fig. 5, the measured and calculated dust loads on snow are presented.

One can see that the data on three points are consistent quite well with each other. The considerable difference at point 1 can be probably accounted for by its location under the crowns of pines (the Rudnichny pine forest)

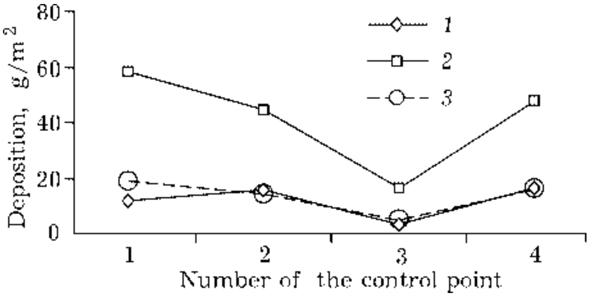

Fig. 5. Comparison of calculated fall-out of dust particles for the winter contained in the emissions of Kemerovo industry with the data of snow analysis. *1* – measurement, *2* – calculation, *3* – calculation (×0.32).

where a part of deposition is retained on the surface of coniferous needles.

It is noteworthy that, unlike the two previous cases, the experimental data on Kemerovo contain information about the dispersion composition of particles in snow samples. It is planned to make a comparison of the measured and calculated values for the dispersed composition in further work after an appropriate updating of the software complex results output unit and, possibly, after estimation of the individual fraction composition of dust emissions, at least for the main sources.

CONCLUSION

A rather simple local model for calculation of long-term (season, year) deposition of particulate matter of industrial sources onto the ground surface has been constructed and realized in software-supported manner. The model is incorporated into a unified regulatory atmospheric pollution program, which permits directly using the standard databases from industrial projects or designs of permissible emission rates are accumulated in the regional Environmental Authority. The model makes it possible to subdivide automatically the total amount of particle emission with respect to their fraction composition depending on the preset number of fractions and the percentage of their contribution. The presence of this possibility and the use of only the available climatic information make it possible to make estimation of deposition in the area of any industrial object or set of objects (included in the standard bases) in real time and with a minimum of additional data handling.

Calculated deposition estimates of industrial dust emissions have been made for three sites of the territory of the Kemerovo Region where the experimental studies of accumulation of suspended substances in the snow cover during winter were carried out. A comparison of experimental results with calculated estimates is presented, which permits concluding that the model reflects rather well (for the given class of problems) the change of quantitative deposition indices for the territory under study. In the cases of Belovo and Kemerovo, where the data of experiments are known in absolute values, the comparison demonstrates a noticeable overstating of modeling results. However, since the causes of the discrepancies may be not only the drawbacks of the model and errors of the data on discharges, but also measurement errors, in order to obtain a reliable conclusion on the quality of the model it is necessary to carry out other representative experiments on snow analysis in the regions of large industrial enterprises.

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