

Experimental and model studies of spatial distribution of the atmospheric aerosol over Lake Baikal

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We discuss results of a complex shipborne research of the spatial distribution of the aerosol fields conducted by means of laser sensing and *in situ* measurements. Based on direct sampling of aerosol followed by chromatographic determination of concentration of individual fractions of polycyclic aromatic hydrocarbons (PAH), we have mapped the concentration distributions over Lake Baikal. It was found that the mean concentrations of the PAH sum were close to the background ones with their spatial distribution being rather nonuniform. Simultaneous analysis of 2-D spatial sections of aerosol fields acquired with lidar has revealed some characteristic tendencies in spatial fluctuations of these parameters. In addition to instrumental investigations, model calculations of the PAH distributions were conducted based on numerical solution of the spatial nonstationary semiempirical equation of the turbulent diffusion of admixtures. The comparison with the experiment has shown the calculated concentrations to be close to the measured ones.

Introduction

To study spatial distribution of aerosol fields, scientists from three institutes of SB RAS have arranged a comprehensive study during “Baikal-02” field mission in July–August 2002 onboard “G. Titov” research vessel of Limnological Institute, SB RAS. Sensing of the atmosphere was carried out in the regions of Southern and Middle Baikal by means of a single-frequency “Loza-M” lidar, accompanied by sampling aerosol, meteorological measurements, and recording the synoptic conditions therein.

In this paper, we present the results of lidar investigations into the spatial structure of aerosol fields based on the data array obtained during shipborne expedition. Determination of 13 PAH (polycyclic aromatic hydrocarbons) included in the list of priority ecotoxicants and recommended for permanent monitoring in the environmental objects was carried out in aerosol samples collected along the survey route. Concentrations of benz(a)pyrene, as the most dangerous carcinogenic substance, from the list of priority PAH, detected in the air over Lake Baikal, are close to the background values in this region. In addition to instrumental observation, calculations were carried out using the mathematical model based on numerical solution of the spatial non-stationary semiempirical equation of turbulent diffusion of admixtures.

1. Instrumented investigations

1.1. Remote technique

Laser sensing of the atmosphere

During the shipborne research mission we have compiled a vast array of data on vertical sounding of the troposphere up to the heights of 5 to 6 km over Lake Baikal. The scheme of routs, conditions of the experiment, and the general synoptic situation were described in detail in our earlier papers.^{1,2}

The sounding was carried out with a “Loza-M”³ single-frequency aerosol lidar operated at the wavelength $\lambda = 0.53 \mu\text{m}$. The lidar return signals operated at a pulse repetition rate of 5 Hz were recorded in the form of one-minute series every 5 minutes. The time and spatial coordinates of each sounding session were recorded, as a separate data file, using a GPS (Global Positioning System) sensor.

Interpretation of the single-frequency aerosol lidar data in the problems of investigation of the vertical structure of aerosol fields under conditions of clean and weakly turbid atmosphere is based on inverting the lidar equation in the single scattering approximation taking into account the height dependence of the molecular scattering in the solution. Reconstruction of the profiles of the scattering coefficient was carried out according to the algorithms of processing for a two-component (aerosol and

molecular) medium, which are based on the Fernald method.⁴ We used the iterative version of the Fernald method,⁵ which adaptively takes into account the functional relation between the total scattering and backscatter in reconstructing the profile of the extinction coefficient from the lidar data. Estimates of the calculated mean error of the method⁵ do not exceed 35% and are the least among the methods generally used for reconstruction of optical characteristics from the data of single-frequency sounding of a weakly turbid atmosphere.

The two-dimensional cross section of aerosol fields over Lake Baikal along the ship route were constructed from the vertical profiles of the optical parameter of the atmosphere $\sigma(H)$ (aerosol extinction coefficient) obtained. Taking into account subsequent averaging within the limits of one-minute measurement sessions and mean velocity of the ship about 14 km/h, the mean section of the profiles $\sigma(H)$ was obtained with the horizontal resolution of 1 km and vertical resolution of 6 m. The upper boundary of the cross section was ~ 3 km and the lower one ~ 0.2 km.

It was revealed from analysis of vertical cross sections of aerosol fields acquired in moving along the coastal line of the lake and across that orographic peculiarities of the region essentially affect the formation of the spatial structure of the field.¹ It was obtained by the methods of correlation analysis of the vertical distribution of the aerosol optical thickness along the ship motion route and the spatial profile of the tops of the Primorskii ridge, that the highest correlation is observed in the lower layer of the atmosphere 0.5–1.5 km.

Qualitative and quantitative estimates of the effect of the coastal landscape were made assuming the geostrophic transfer of air masses determined by synoptic conditions. So, the apparatus of continuous wavelet transformation was used for a more detailed analysis of the sections of the aerosol field. It enabled us to reveal characteristic frequency scales and trends in the spatial fluctuations of $\sigma(H)$.² Let us present here only main conclusions drawn in Ref. 2. The height level, above and below of which the behavior of the wavelet coefficients is different, is close to the value of the correlation height, which can be taken equal to the height of the mixing layer. The effect of the mountain ridge on the structure of aerosol field manifested itself in the formation of leeward waves and it is stronger in the middle part of the lake between higher mountains. Here the wave motions of different spatial scales are observed in the aerosol field along the entire routes between the lakesides.

The small-scale perturbations formed behind the ridge in the southern part of the lake propagate at a insignificant distance from the coast and fade by the middle of the lake. The characteristic peculiarity of the low-frequency atmospheric processes is the presence of linear frequency trend (decrease of frequency of oscillations when moving from west to east).

Then, for comparative analysis of the two-dimensional aerosol distribution (obtained from the lidar data) and the one-dimensional distribution of

PAH concentrations (obtained from measurement data after contact aerosol sampling) over the lake, the AOT (aerosol optical thickness) of the 1.5-km high atmospheric layer was considered. It most completely represents the fluctuations of the boundary layer of the atmosphere related with the effect of the coastal relief on the trans-boundary transfer of industrial atmospheric admixtures, the tracer of which is atmospheric aerosol. The map of spatial distribution of AOT averaged over all days of the lidar survey with coordinate reference over the water area of Lake Baikal (Fig. 1) was constructed using the Surfer Mapping System⁶ package.

The conclusions on the main tendencies of the aerosol spatial fluctuations drawn earlier^{1,2} were taken into account in constructing the map, that allows us totally and, mainly, adequately present the spatial variations of the vertical profiles of the optical thickness for individual routes across the lake even under conditions of inhomogeneous and not complete coverage of the area of Lake Baikal with measurements.

It is seen in Fig. 1 that quite homogeneous filling of the southern part of the Lake Baikal with aerosol is observed, as well as the scales of variations of the optical thickness characteristic of the high-mountain part of the middle Baikal (near Ol'khon island). Besides, the local areas of enhanced values (relative to the mean background level) of the optical thickness were revealed. It is the section along the direction toward the head of Angara river and in the middle of the lake in the section from cape Krestovski to Oblom cape. It should be noted that the values of the background AOT (they are presented in our estimates with subtraction of the contribution due to molecular scattering) of a 1-km high layer of the atmosphere do not exceed 0.01 under conditions when the meteorological visibility range over the lake reached 200 km. One of the sources of increasing atmospheric turbidity over the Lake Baikal can be the admixtures of anthropogenic origin.

1.2. Contact method

Local sampling of aerosol.

Technique for determining the concentrations of separate fractions of PAH in aerosol samples

Polycyclic aromatic hydrocarbons take especial place among anthropogenic admixtures polluting the atmosphere, soil, and natural waters. The PAH in the atmosphere are mainly sorbed on solid aerosol particles. Determination of 13 PAH was carried out in the aerosol samples collected along the survey routes.

Aerosol samples were collected on "Whatman EPM 2000 High purity glass microfibre filters" glass fiber filters of the size 17×21 cm by means of PM-10 pumps by "Andersen Samplers, Inc." at a flow rate of 2 m³/min. The volume of air pumped through the filter was 200 to 650 m³. The sample collector was installed in the ship rostrum at the height of 10 m above the water.

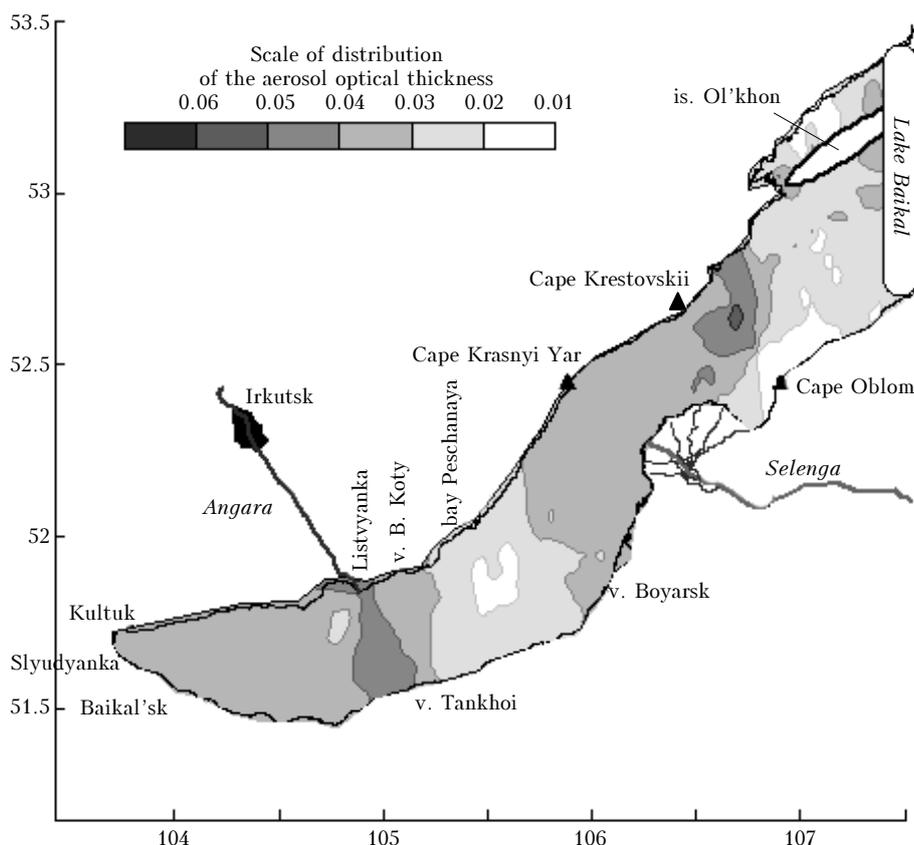


Fig. 1. Map of the spatial distribution of the aerosol optical thickness of the 1.5-km high atmospheric layer over Lake Baikal. Coordinate axes represent the degrees of northern latitude (vertical) and eastern longitude (horizontal).

The fraction of PAH was selected from aerosol by thrice-repeated extraction with acetonitrile in an ultrasound bath at room temperature. The obtained extract was concentrated to the volume of 0.1–0.2 ml and then was chromatographed by means of a “Agilent, GC 6890, MSD 5973 Network” chromatomass-spectrometer under the following conditions: the 50 m long capillary column with the internal diameter of 0.32 mm, the BetaBasic SE 30 (SE-54) phase with the efficiency not less than 100000 theoretical plates; the injector temperature 280°C, the column temperature 50°C during 5 minutes, then the temperature gradient of 15°C per 1 min in the range from 50 to 300°C and then 300°C during 15–30 min.

The peaks of PAH in the chromatograms were identified according to the times of retaining and according to the mass-spectra in the data library. Quantitative measurements of PAH were carried out using the internal standards of “Supelco” (USA): phenantrene d_{10} , chrisen d_{12} , and perylene d_{12} . The error in measurements did not exceed 10%. The data obtained are presented in Table 1.

The map (Fig. 2) of spatial distribution of the PAH concentrations over Lake Baikal was constructed using all the obtained data on the total values of PAH. The maximum concentrations of the total amount of identified PAH are characteristic of the Southern Baikal in the regions of the city of

Baikalsk and Tankhoi village, and that of middle Baikal are observed in the region of Oblom cape.

Benz(a)pyrene is related to the most dangerous carcinogen from the list of priority PAH. It is recommended in Russia for permanent monitoring in the environmental objects. Concentrations of benz(a)pyrene (1–64 $\mu\text{g}/\text{m}^3$) detected in the air over Baikal are close to the background values for the region (20–100 $\mu\text{g}/\text{m}^3$) observed over the territory of Barguzin national park.⁷

In identifying the sources according to the relative concentrations of PAH in the atmosphere, as well as at monitoring of the environment, one should take into account relative stability of PAH. According to Ref. 8, the most reactive-capable PAH is benz(a)pyrene, and the most inertial is benz(e)pyrene. The sources of combustion emit both isomers in equal amounts. In aerosol samples collected near the sources of pollution the ratio

$$R = \text{benz(e)pyrene} / [\text{benz(e)pyrene} + \text{benz(a)pyrene}]$$

is equal to 0.5–0.57.^{9,10} The concentration of benz(a)pyrene decreases during the process of photochemical destruction, and the ratio R changes within the limits from 0.70 to 0.83 in aerosol samples collected at significant distances from the sources of PAH.^{11,12}

Table 1. Range of the concentrations of PAH in aerosol (pg/m³) over Lake Baikal in July–August 2002

Place and date of aerosol sample collection	Phenanthrene	Anthracene	Pyrene	Fluoranthene	Benz(a)-anthracene	Chrisen	Benz(b)-fluoranthene	Benz(k)-fluoranthene	Benz(e)-pyrene	Benz(a)-pyrene	Perylene	Indeno-(1,2,3c,d)-pyrene	Dibenz(a,h)-anthracene	Benz(g,h,i)-perylene	Sum of PAH
v. Bol'shie Koty, 07.28.02	25.7	4.6	5.7	15.0	2.9	2.2	4.6	4.5	3.6	4.5		18.0	3.8	18.0	113.0
b. Peschanaya, 07.28–29.02	75.3	9.4	11.1	25.4			1.1	1.2	0.7	0.8		2.0		1.6	128.5
b. Peschanaya – Boyarsk, 07.29.02	114.7	18.8	20.3	56.2			2.5	3.0	2.2	2.9		17.5	3.2	18.8	260.1
Boyarsk – Listvyanka, 07.29.02	18.8	4.2	5.9	11.3	1.7	1.4	1.8	1.4	1.1	1.3		3.4		2.4	54.7
v. B. Koty – Goloustnoe, 07.30.02	44.9	3.9	4.6	1.4											54.8
v. Bugul'deika – b. Aya, 07.30.02	64.5	9.8	10.9	37.6											122.8
c. Oblom, 07.30.02	313.6	42.3	52.9	277.2											686.0
c. Khoboi – c. Khuzhir, 07.31.02	3.4	6.4	14.0	32.8		4.7									61.3
Strait Ol'khonskie vorota – c. Krasnyi Yar, 07.31.02	86.2	7.2	73.1	77.5	3.5	39.1	33.2	13.0	17.8	6.0		26.4	5.2	20.3	408.5
c. Krasnyi Yar – middle of the Lake, 08.01.02	63.7	12.9	79.1	136.4	5.3	27.6	46.9	17.3	25.8	9.4		41.9	7.8	33.5	507.8
v. Tankhoi – Listvyanka, 08.01.02	100.3	11.5	43.8	54.2	14.1	25.1	46.7	21.9	27.9	16.4	5.5	68.7	17.7	67.0	520.8
v. B. Koty – Baikalsk, 08.01.02	52.7	8.8	15.5	37.3	4.2	31.0	10.1	5.9	6.7	4.4		9.3		8.3	194.4
Baikalsk, 08.01.02	30.9		12.5	12.7	12.5	7.5	23.5	23.0	22.0	29.0	9.5	195.7	39.5	224.8	643.0
Baikalsk – v. Tankhoi, 08.02.02	30.9	4.8	17.6	23.7	2.9	7.6	12.0	5.9	6.9	3.9	1.0	18.4	4.3	16.4	156.1
Goloustnoe – Baikalsk, 08.01.02	44.0	4.3	28.5	34.7	2.6	10.7	15.4	6.3	8.3	3.8		9.9		7.0	175.5
Baikalsk, 08.02.02	98.9	12.2	69.7	51.7	11.6	24.5	47.8	29.1	27.9	25.0		99.3	22.1	92.1	611.9
c. Posol'skii sor – v. Tankhoi – v. B. Koty, 08.31.02	33.9	5.0	14.9	39.3	2.2	5.0	7.1	0.4	3.9	3.5		6.3		4.9	126.3
v. B. Koty – b. Peschanaya, 08.03.02	37.5	7.0	8.4	21.1	1.9	2.1	2.4	1.8	1.3	1.9				3.1	88.6

The calculated ratios R in air over the region of Lake Baikal change from 0.43 to 0.76 depending on the distance between the site of collection and the source of pollution (Table 2).

Table 2. Concentration of benz(a)pyrene normalized to the sum of concentrations of benz(e)pyrene and benz(a)pyrene (R) over Lake Baikal in July–August 2002

Region of sampling	R
Baikalsk	0.43
b. Peschanaya	0.45
v. Boyarsk	0.48
v. Bol'shie Koty	0.54
section Baikalsk – v. Tankhoi	0.64
section v. Tankhoi – Listvyanka	0.63
section Ol'khonskie vorota – c. Krasnyi Yar	0.75
section c. Krasnyi Yar – middle of the lake	0.73
section c. Krestovskii – c. Oblom	0.76

Individual R values are shown in Fig. 2 at the middle points of the route of collecting the aerosol samples. As is seen, according to above said, the characteristic small R values were obtained at costal sites of Southern Baikal, the sites extremely close to

the source in Baikalsk, and intermediate values are observed in the region of village Tankhoi. The maximum concentrations of PAH in the region of Oblom cape, according to the values R , are characteristic of the most remote sources.

2. Model investigations

2.1. Calculations by a mathematical model

In addition to instrumented research, calculations were made using a mathematical model based on numerical solution of spatial semiempirical equation of turbulent diffusion of admixtures.¹³ Dispersal of the main PAH emitted by stationary sources and motor transport of Irkutsk, Slyudyanka, Baikalsk, Kamensk, and Selenginsk was considered. The sources of emission of PAH were also motor and railway transport on the route from Irkutsk to Ulan-Ude. Estimation of the mass flow rate of PAH was made based on the results from Refs. 14–19. Statistical characteristics of the wind field used in calculations were obtained from processing the data of long-term observations of the vector of wind velocity.^{20,21}

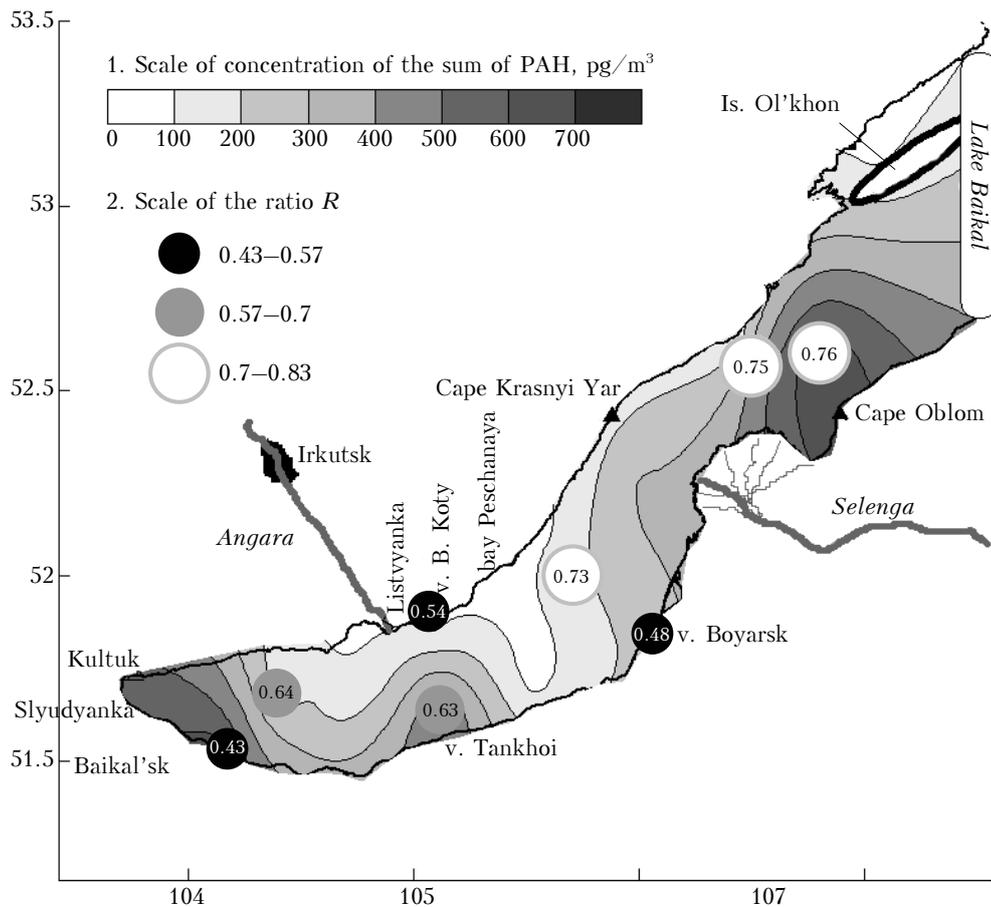


Fig. 2. Scale 1 is spatial distribution of the concentration of the sum of PAH, pg/m^3 ; scale 2 is concentration of benz(e)pyrene normalized to the sum of concentrations of benz(e)pyrene and benz(a)pyrene at the middle point of the route of aerosol sampling.

Concentrations of the sum of 13 PAH were calculated within the area of 500×250 km and the height of 5.5 km over the surface of Lake Baikal. The temporal and horizontal steps were, respectively, 150 s and 5 km, the vertical step was set as follows: it was equal to 50 m up to the height of 600 m, then 200, 200, 1000, 1500, and 2000 m. The coefficients of turbulent diffusion were calculated using the relationships of the semiempirical theory of turbulence.¹³

Some results of the first series of numerical experiments are shown in Fig. 3a. The maximum calculated concentrations of PAH in the regions of Oblom cape and near the city of Slyudyanka are of the same order that the measured ones (see Fig. 2). The calculated concentrations of PAH in the region of Tankhoi are low (less than $100 \text{ pg}/\text{m}^3$).

In order to explain great values of the measured concentrations of the sum of PAH in the region of village Tankhoi, the following series of numerical experiments was carried out. It was assumed that there is a source of emission of PAH in the region of Tankhoi, the mass flow rate of which is $25 \text{ mg}/\text{s}$. One of the calculated results is shown in Fig. 3b. It is seen that the configuration of isolines in Fig. 3b is

similar to the configuration of isolines in Fig. 2. Further comprehensive investigations are necessary for confirmation (or disproof) of the hypothesis about a powerful source of PAH in the region of village Tankhoi.

2.2. Analysis of back trajectories of the air mass transfer

Similar tendencies are observed in analysis of back trajectories of the air masses transfer to the regions under study (Fig. 4). Additional consideration of the sections of the vertical flows at different levels along the calculated trajectories enables one to determine the probability image of the drift of anthropogenic aerosol admixtures, the indicator of which is PAH.

Thus, the most probable for the region of Oblom cape is the trajectory at the level of the boundary layer (1.5 km) (Fig. 4a), transferring admixtures by downward flows to the region of measurements.

Air masses are transferred from distances of hundreds kilometers: they pass over the transport path at the eastern coast of Lake Baikal and inhabited

area situated there, and are replenished from the mixing layer (500 m) by upward flows. Less probable is the drift of admixtures by airflows from the level of the free atmosphere (3000 m) from the directions of the northwest coast of Baikal, because upward flows are detected along the entire trajectory, which lead to possible formation of clouds as was observed in synoptic conditions of that period. Let us also note the intermittent of upward and downward flows at the level of the top of the boundary layer, that was observed earlier^{1,2} when analyzing the distribution of the spatial structure of the optical thickness depending on the synoptic conditions and the effect of coastal relief of mountains.

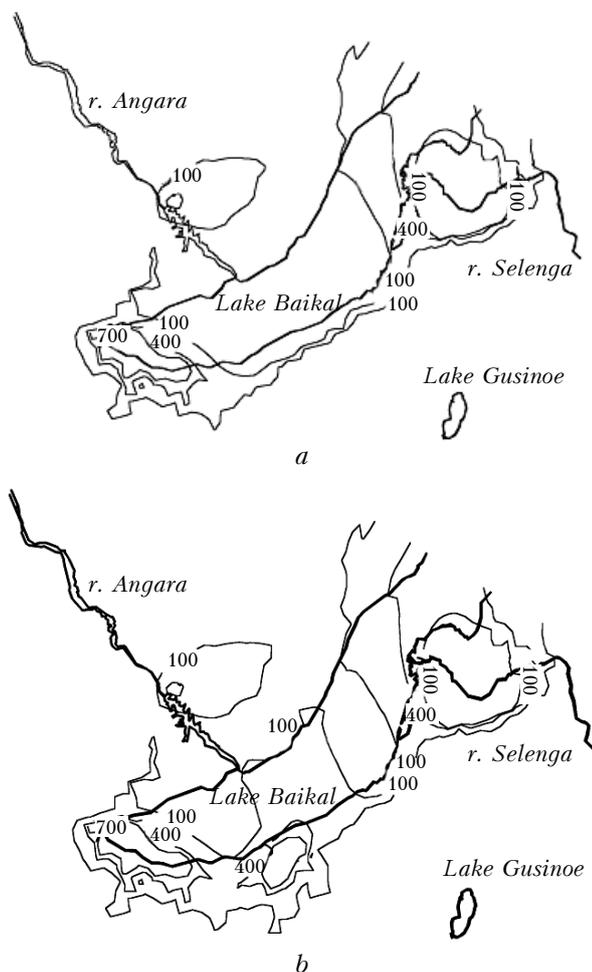


Fig. 3. Isolines of the calculated mean near-ground concentrations (pg/m^3) of the sum of PAH in the region of Lake Baikal (a); the same, but with the source of emissions in the region of village Tankhoi (b).

Analysis of temperature sections and the sections of vertical motions along the back trajectories in the direction to Tankhoi village (Fig. 4b) shows quite stable stratification of the atmosphere in the boundary layer during the measurements. Insignificant scaled variations of the vertical flows are caused mainly by the diurnal behavior of temperature and do

not go out of the limits of the boundary layer. That means, that the main contribution from anthropogenic admixture comes along the trajectory of the near-ground level (100 m), and less significant contribution comes along the trajectory at the level of the mixing layer (500 m). According to these heights, the distance of the transfer is determined by no more than tens of kilometers and is caused by local circulation inside the basin. The main source of anthropogenic admixture, the indicator of which are the revealed enhanced values of the concentration of the sum of PAH along the trajectories of the near-ground layer, is situated in the coastal zone of Tankhoi village. The additional source is the atmospheric channel corresponding to the trajectory of transfer from northwest direction along the head of Angara river, that is also confirmed by the map of distribution of the optical density of the incoming aerosols (see Fig. 1).

Thus, the calculated ratios R over the Lake Baikal directly depend on the distance between the site of sample collection and the source of pollution.

Conclusions

It has been shown that the recorded mean total concentration of the priority PAH in the air over Lake Baikal are close to the values determined in the background regions. Nevertheless, the spatial distribution is quite inhomogeneous. The places with both lower and higher concentrations are observed, that is seen in the spatial maps of distribution of PAH in the atmosphere over Lake Baikal.

The values R calculated for the region over Lake Baikal adequately correspond to the dependence of the distance between the site of sample collection and the source of pollution. The values R within the limits from > 0.65 (the region of Southern Baikal) to < 0.75 (the region of middle Baikal) correspond to mesometeorological scale of atmospheric motions at the distance from several tens kilometers (> 10 km) to several hundred kilometers (< 300 km).

Analysis of the maps of the spatial distribution of AOT (see Fig. 1), PAH (see Fig. 2), synoptic conditions, and the maps of back trajectories (see Fig. 4) of the air transfer in the region of investigation show the characteristic tendencies in the spatial fluctuations of these parameters. Owing to this fact, one can identify (taking into account the distribution of the estimate of R) the sources of incoming aerosol admixtures transported to Lake Baikal through the atmospheric channel during trans-boundary transfer of the air masses.

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