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Physical characteristics of concentration fields of tropospheric bioaerosols in the South of Western Siberia

Alexander I. Borodulin[a,*], Alexander S. Safatov[a], Alexander N. Shabanov[a], Andrei A. Yarygin[a], Olga G. Khutorova[b], Boris D. Belan[c], Mikhail V. Panchenko[c]

[a] *Department of Ecological Aerobiology, State Research Center of Virology and Biotechnology "Vector", Koltsovo, Novosibirsk Region 630559, Russia*
[b] *Kazan State University, Kazan, Russia*
[c] *Institute of Atmospheric Optics SB RAS, Tomsk, Russia*

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Abstract

The State Research Center of Virology and Biotechnology "Vector" and the Institute of Optics SB RAS are performing systematic study of tropospheric bioaerosols in the South of Western Siberia. The work summarizes the results of analysis of some physical characteristics of the data array on bioaerosol concentration obtained with an airplane laboratory at the altitudes of 0.5, 1, 1.5, 2, 3, 4, 5.5 and 7 km. The flights were performed during the last 10 days of each month in 1999–2003. It was shown that the concentration of total protein aerosols obeyed the laws of continual statistics, and the concentration of culturable microorganism aerosols obeyed the laws of discrete statistics. The analysis of correlation coefficients and cross correlation of bioaerosol concentration fields was performed. Wavelet analysis of the data showed that variations of tropospheric bioaerosol concentration were mainly determined by typical seasonal processes with periods of 12, 6, 4 and 8–9 months. Seasonal variations cause approximately 80% of the total dispersion of variations of total protein concentration, and the amplitudes of variations of culturable microorganism concentration are small as compared with the constant component. Harmonic analysis of bioaerosol concentration fields was performed. Harmonics that are significant in wavelet spectra for all time series were taken for the approximation of the series. It was revealed that four-month periodicity was internal and was caused by a wave spreading from above with a vertical rate of about 4 km/month.

Keywords: Bioaerosols; Troposphere; Total protein; Culturable microorganisms; Concentration fields

* Corresponding author. Tel.: +7 383 2 367 469; fax: +7 383 2 367 409.
E-mail address: borodulin@vector.nsc.ru (A.I. Borodulin).

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1. Introduction

The State Research Center of Virology and Biotechnology "Vector" and the Institute of Optics SB RAS are performing a systematic study of tropospheric bioaerosols in the South of Western Siberia. Our work investigates aerosols containing total protein and culturable microorganisms.

Sampling is performed with "Optic-E" laboratory mounted on an Antonov-30 airplane during the last 10 days of each month. The lane of 50 km passes over Karakan forest located on the right bank of the Ob River. Coordinates: longitude 54°30′ North; latitude 82°20′ West. The airplane flies along this lane in the daytime successively over a forestland at the altitudes of 7, 5.5, 4, 3, 2, 1.5, 1 and 0.5 km. To analyze total atmospheric protein, an air sample is collected for each altitude onto fibrous filters of HEPA type for 10–20 min. The collected air volume is approximately 2 m^3 per filter. To detect culturable microorganisms, air samples are collected on impingers (Griffiths & DeCosemo, 1994) filled with 50 ml of physiological solution at the flow rate of 50 l/min.

Total protein content is analyzed in a laboratory with one of the two methods: first—according to the Bradford method (Darbe, 1989); the method sensitivity is 0.1 μg/ml, and the error of the measured concentration value does not exceed 30%; second—according to the fluorescent method using a dye, see (You, Haugland, Ryan, & Haugland, 1997); the method sensitivity is 0.1 μg/ml of the sample, and the error of the measured concentration value does not exceed 20%.

To detect culturable microorganisms, the collected samples are seeded onto Petri dishes containing the following agar media: complete LB medium (Darbe, 1989) and depleted LB medium (diluted 1:10) for detection of saprophytic bacteria; starch-ammoniac medium SAM (Saggie, 1983) for detection of actinomyces; soil agar, Sabourau's and Czapek's media (Saggie, 1983) for detection of lower fungi and yeasts. Serial dilutions of the samples are prepared when needed. The seeded samples are incubated in a thermostat at 30 °C for 3–14 days. The morphological characteristics of the detected microorganisms are studied visually and by light microscopy. To do this, fixed Gram-stained cell preparations and live preparations of cell suspensions are prepared and observed by the phase contrast method. The taxonomic groups the microorganisms belong to are determined up to the Genus (Starr, Stolp, Truper, Balows, & Schlegel, 1981; The Methods of Experimental Mycology, 1982). Standard techniques are employed to calculate the numbers of culturable microorganisms in the samples (Ashmarin & Vorobyev, 1962), in which the numbers of microorganisms is averaged over 2–3 parallel samples distributed on 4–5 different media.

Thus, a sufficiently large array of data on bioaerosol concentrations in the troposphere of the South of Western Siberia has been accumulated during the observation period from 1999 to 2003: 2×8 time series of 60 readings each are available. The concentrations of total protein aerosols are given in micrograms per cubic meter, and the concentrations of live microorganisms aerosols are given in number per cubic meter. These are the mass and the countable concentrations, respectively.

Some preliminary results of analysis of the obtained experimental data are published in the works (Andreeva et al., 2000, 2001; Ankilov et al., 1999, 2001; Belan, 2000; Borodulin et al., 2003b; Safatov et al., 2003). The results are summarized in the work (Andreeva et al., 2002). In particular, the analysis of the obtained data showed that:

1. There is a clear seasonal variation of tropospheric bioaerosol concentration associated with the change of the seasons;
2. No obvious relation is observed between bioaerosol concentration fields and the altitude;

3. There is a considerable distribution in concentration values, which exceeds the estimated errors of measurement.

On this basis we concluded that concentration fields of tropospheric bioaerosols in the South of Western Siberia are mainly determined by remote pollutant sources and their structure is significantly influenced by the statistical nature of the process of aerosol pollutants spread in atmosphere.

The present work contains some results of analysis of physical characteristics of bioaerosol concentration fields: the type of distribution functions of bioaerosol concentration has been determined; correlation coefficients and cross-correlation coefficients of bioaerosol concentrations have been calculated; wavelet and harmonic analyses of the available data series have been performed; examples of approximation of concentration series with harmonic series are given.

2. The statistics of concentration of atmospheric bioaerosols

Proceeding from the general considerations, it can be supposed that the fraction of tropospheric aerosol containing total protein is formed of particles having a wide size range and contains a large number of macromolecules with common specific fragments. In extreme case, the mechanism of the formation of protein component of tropospheric aerosol may be considered the result of successive splitting of some sufficiently large "initial" particles. According to Kolmogorov's theorem (see its discussion in Fuchs, 1964), such a system must be described in the limit by the laws of continual statistics, namely by logarithmically normal distribution. The component of atmospheric aerosol containing culturable microorganisms is presented with an ensemble of indivisible subunits. Destruction or death of a microorganism removes it from the list of the living. Thus, it should be supposed that culturable microorganism faction in tropospheric aerosol must be described with the laws of discrete statistics, and total protein faction must be described with the laws of continual statistics.

The logarithmically normal distribution function looks as follows:

$$F(C) = \frac{1}{2}\left[1 + \operatorname{erf}\left(\frac{\ln C - M}{\sqrt{2}\Sigma}\right)\right], \tag{1}$$

where C is the argument of the distribution function; $\operatorname{erf}(\ldots)$ is the probability integral. The parameters of the distribution law M and Σ are expressed via the expected value of the concentration $\overline{C}$ and its dispersion σ^2 as follows:

$$\overline{C} = \exp\left(M + \frac{\Sigma^2}{2}\right), \quad \sigma^2 = \exp(2M + \Sigma^2)[\exp(\Sigma^2) - 1]. \tag{2}$$

The process of spread of atmospheric aerosols is characterized by the presence of the effect of the concentration intermittence, i.e. the probability of observing its zero values. We can help understand it by visualizing the behavior of smoke from a fire or the spread of puffs of smoke from chimneys of plants. The distribution function (1) does not take this effect into account. That is why we will consider another continual distribution function of atmospheric pollutant concentration (Borodulin, Maistrenko, & Chaldin, 1992), which does take into account the presence of the effect of the concentration

intermittence,

$$F(C) = 1 + \frac{1}{2}\left[\operatorname{erf}\left(\frac{C-\overline{C}}{\beta}\right) - \operatorname{erf}\left(\frac{C+\overline{C}}{\beta}\right)\right], \tag{3}$$

where β is the second parameter of the distribution function. Expression (2) is an exact analytical solution of the Focker–Plank–Kolmogorov equation obtained under the assumption that the random process of variation of atmospheric pollutant concentration in the given point of space is a Markovian one. It is convenient to determine the parameter β using the relationship

$$\frac{\sigma^2}{\overline{C}^2} = \operatorname{erf}(\beta_0)\left(1 + \frac{1}{2\beta_0^2}\right) - 1 + \frac{1}{\sqrt{\pi}\beta_0}\exp(-\beta_0^2);\ \beta_0 = \frac{\overline{C}}{\beta}. \tag{4}$$

Discrete statistics of the atmospheric pollutant concentration was considered in Borodulin, Desyatkov, and Kotlyarova (1997), where the binomial distribution for k particles in the given volume was substantiated based on natural assumptions. If the particle number is large enough (in our case, the order of magnitude of the culturable microorganisms number in $1\,\mathrm{m}^3$ makes several hundreds or thousands), the binomial distribution is approximated by Poisson distribution

$$p(k) = \exp(-\overline{k})\frac{(\overline{k})^k}{k!}, \tag{5}$$

where $p(k)$ is the probability of observing k particles in a unit of volume; $\overline{k}$ the expected value of particle number.

It has been already said that total protein and culturable microorganism concentrations on the average practically do not depend on the flight altitude. That is why the ensemble of experimental data on total protein concentration C_p and culturable microorganism concentration C_b was treated as follows. First total protein concentrations $C_{\mathrm{p}m}$ and culturable microorganisms concentrations $C_{\mathrm{b}m}$ averaged over altitudes of observations h were found for each experiment. Then the values $\varphi_\mathrm{p} = C_\mathrm{p}/C_{\mathrm{p}m}$, $\varphi_\mathrm{b} = C_\mathrm{b}/C_{\mathrm{b}m}$ were calculated, and standard deviations of normalized concentrations of total protein $\sigma_{\varphi_\mathrm{p}}$ and culturable microorganisms $\sigma_{\varphi_\mathrm{b}}$ averaged over the whole ensemble of performed experiments were determined. The ensemble of total protein concentrations was presented with $n = 245$ experiments, and the ensemble of culturable microorganism concentrations with $n = 197$ experiments.

The characteristic feature of Poisson distribution is that its dispersion is equal to the expectation $\sigma_k^2 = \overline{k}$. This proportion may be proposed as a hypothesis about the correspondence of distribution functions of total protein and culturable microorganism concentrations to Poisson statistics. According to the employed method of treating experimental data, the values φ_p and φ_b are proportionate to the particle number in the sample, and their expected values equal 1. Therefore, in our case, it is necessary to test the hypotheses: $\overline{\sigma^2_{\varphi_\mathrm{p}}} = 1$ and $\overline{\sigma^2_{\varphi_\mathrm{b}}} = 1$ where the line means the averaging-out over the ensemble of experimental data. These values as well as the values of statistics

$$T_\mathrm{p} = \frac{\overline{\sigma^2_{\varphi_\mathrm{p}}} - 1}{S_\mathrm{p}/\sqrt{m}} \text{ and } T_\mathrm{b} = \frac{\overline{\sigma^2_{\varphi_\mathrm{b}}} - 1}{S_\mathrm{b}/\sqrt{m}},$$

which must have Student's t distribution with the number of degrees of freedom $m = 8$ corresponding to eight altitudes of concentration measurement are presented in Table 1. Standard sample deviations S_p,

Table 1
Testing of hypothesis about the equality of dispersion and expected value of the particle number

	Total protein	Culturable microorganisms
Expected values averaged over an ensemble of observations, $\sigma^2_{\varphi_p}$ И$\sigma^2_{\varphi_b}$	$\overline{\sigma^2_{\varphi_p}} \pm S_p = 0.29 \pm 0.32$	$\overline{\sigma^2_{\varphi_b}} \pm S_b = 2.24 \pm 1.83$
Statistics T_p and T_b:	$T_p = -6.28$	$T_b = 1.92$
Fractile Student's t distribution $\lvert t_m \rvert_{1-\alpha}$; $\alpha = 0.05$, $m = 8$:	2.31	

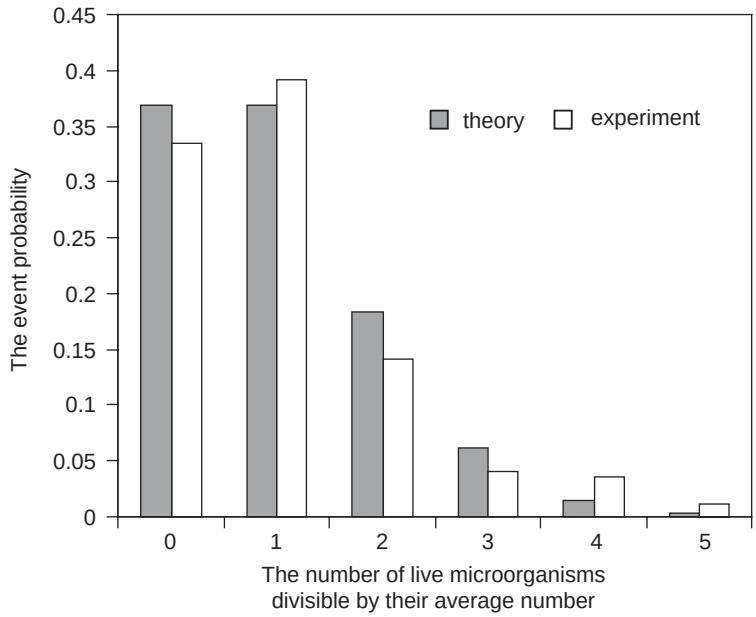

Fig. 1. Theoretically and experimentally determined probabilities that the number of microorganisms in a sample is a divisible by their average number.

S_b of the values $\overline{\sigma^2_{\varphi_p}}$, $\overline{\sigma^2_{\varphi_b}}$ and fractiles of Student's t distribution $|t_m|_{1-\alpha}$ for $\alpha = 0.05$ and $m = 8$ are also given there. The module of statistics value T_p exceeds the given fractile, and the statistics T_b are lower. We see that the proposed hypothesis is discarded for the distribution of total protein concentration and is accepted for the distribution of culturable microorganism concentration. Testing of this hypothesis is a necessary but not a sufficient condition for concluding that the law of distribution of culturable microorganism concentration refers to Poisson statistics. That is why direct testing of the correspondence of the ensemble of data to the statistics Eq. (5) is especially interesting. In the general case, the considered value φ_b is not integral. However, appropriate evaluation is still possible. Supposing that $k = 0, \bar{k}, 2\bar{k}, \ldots$ and as $C_b = k/V$, where V is the considered sample volume, we will obtain a series of integral values $\varphi_b = 0, 1, 2, \ldots$ It corresponds to the number of microorganisms in $1\,m^3$, a multiple of their average number. The distribution of the φ_b value should also obey Poisson statistic. Fig. 1shows the probabilities $p(\varphi_b)$ calculated according to Eq. (5) and sample frequencies of occurrence of the given values φ_b found

Table 2
Testing of hypothesis about the distribution of culturable microorganisms number multiple of the average value according to Poisson law

φ_{b_i}	Frequency of occurrence of events, h_i	$p(\varphi_{b_i})$	$np(\varphi_{b_i})$	$\chi_i^2 = \frac{[h_i - np(\varphi_{b_i})]^2}{np(\varphi_{b_i})}$
0	66	0.37	72.5	0.58
1	77	0.37	72.5	0.28
2	28	0.18	36.3	1.90
>3	8	0.08	15.6	3.70
			Statistics: $\chi^2 = \sum_i \chi_i^2 = 6.46$	

Distribution fractile $\chi^2_{m,1-\alpha}$; $\alpha = 0.05$, $m = 3$: 7.81

with the ensemble of the conducted experiments. As there are always errors of measurements, it is not always possible to pick out strictly zero concentration values in the available sampling φ_b. That's why we preset the interval $0 \leqslant \varphi_b \leqslant 0.2$ when evaluating the probability of occurrence of zero values. Table 2 contains the results of testing this hypothesis. The first column gives the intervals of φ_b values. The second one contains sample frequencies of occurrence of this event h_i. The third one presents the theoretical data on the probability of occurrence of this event. The fourth one contains the components of statistics χ_i^2. It can be seen that the statistics value χ^2 is lower than the distribution fractile $\chi^2_{m,1-\alpha}$ for $\alpha = 0.05$ and $m = 3$. It means that the hypothesis about the correspondence of this distribution to Poisson statistics is accepted with confidence probability $1 - \alpha = 0.95$.

The supposition about the continuity of the law of distribution of atmospheric protein concentration is an alternative hypothesis to Poisson statistics considered above. Figs. 2a and b present empiric histograms of distribution functions of total protein concentration $F(\varphi_p)$ and culturable microorganism concentration $F(\varphi_b)$ constructed on the basis of an ensemble of conducted experiments. The distribution functions (1) and (3) are given there, too. According to the employed method of data processing $\overline{\varphi}_p = \overline{\varphi}_b = 1$. Table 1 presents dispersions averaged over an ensemble of data $\overline{\sigma^2_{\varphi_p}}$ and $\overline{\sigma^2_{\varphi_b}}$. The parameter β_0 was determined with these values by solving Eq. (4). For a sampling of total protein concentrations $\beta_0 = 1.3$ was obtained, and for a sample of culturable microorganism concentrations—$\beta_0 = 0.36$. The parameters of the logarithmically normal distribution law (1) calculated according to Eq. (2) for an array of data on total protein concentration are $M = -0.13$ and $\Sigma = 0.50$. For culturable microorganism concentration their values are $M = -0.59$ and $\Sigma = 1.08$. It can be seen that the empiric distribution function of total protein concentration qualitatively corresponds to continual distribution functions (1) and (3). Grouping of data enables to satisfy the criterion χ^2 for a logarithmically normal distribution at the significance level $\alpha = 0.01$ (1). These results are shown in Table 3. The proposed hypothesis at the significance level $\alpha = 0.005$ and grouping of data enables to the criterion χ^2 also to be satisfied for the distribution law (3). These results are shown in Table 4. Thus, the hypotheses that the distribution function of total protein concentration corresponds to the laws (1) and (3) is accepted with the confidence probability $1 - \alpha \approx 0.99$–0.995. Contrary to the above, the empiric distribution function of culturable microorganism concentration even qualitatively does not correspond to continual laws of distribution (1) and (3), see Fig. 2b.

The logarithmically normal distribution law considered above strongly corresponds to the zero probability of observing zero concentration values $F(0) = 0$. Contrary to this, the probability of observing

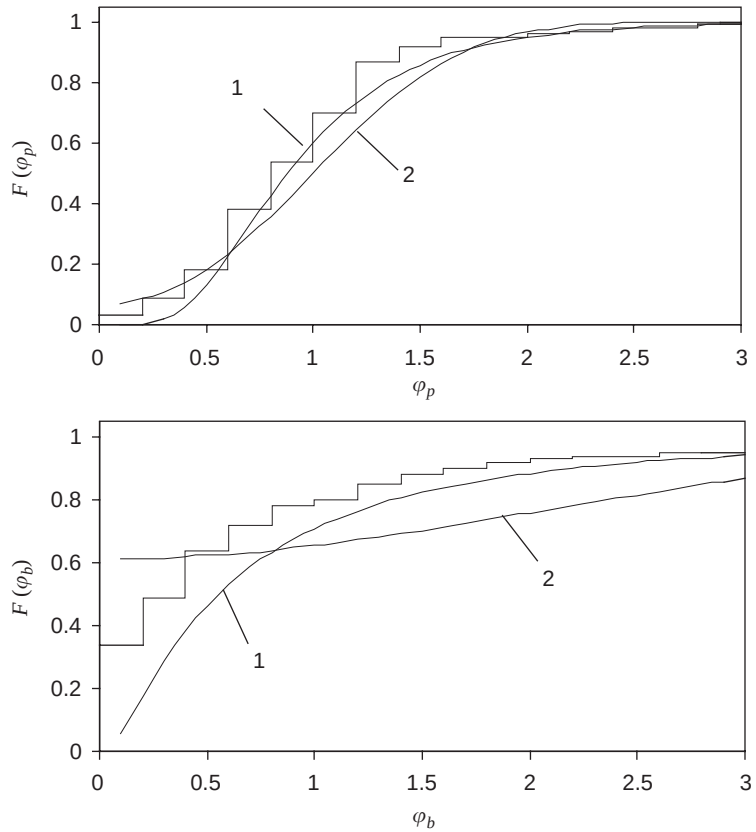

Fig. 2. Empirical distribution functions of total protein concentration and live microorganism concentration (histograms) as compared with distribution functions (1) И (3), curves 1 and 2, respectively.

Table 3
Testing of hypothesis about the distribution of total protein concentration according to logarithmically normal law

$\varphi_{\mathrm{p}i}$	Frequency of occurrence of events, h_i	$p(\varphi_{\mathrm{p}_i})$	$np(\varphi_{\mathrm{p}_i})$	$\chi_i^2 = \frac{[h_i - np(\varphi_{\mathrm{p}_i})]^2}{np(\varphi_{\mathrm{p}_i})}$
0–0.4	21	0.06	18.48	0.34
0.4–0.8	71	0.37	89.43	3.80
0.8–1.2	80	0.31	74.97	0.34
1.2–1.8	60	0.19	46.55	3.89
> 1.8	13	0.08	19.11	1.95
			Statistics: $\chi^2 = \sum_i \chi_i^2 = 10.32$	
Distribution fractile $\chi^2_{m,1-\alpha}$; $\alpha = 0.01$, $m = 3$: 11.34				

zero concentration values for the distribution law (3) is $F(0) = 1 - \mathrm{erf}(\beta_0)$. In the available array of experimental data we have two strictly zero concentration values $\varphi_{\mathrm{p}} = 0$ and four values that are sufficiently close to zero $0 < \varphi_{\mathrm{p}} \leqslant 0.1$. Thus, the frequency of occurrence of "zero" concentration values

Table 4
Testing of hypothesis about the distribution of total protein concentration according to law (3)

$\varphi_{\mathrm{b}i}$	Frequency of occurrence of events, h_i	$p(\varphi_{\mathrm{p}i})$	$np(\varphi_{\mathrm{p}i})$	$\chi_i^2 = \frac{[h_i - np(\varphi_{\mathrm{p}i})]^2}{np(\varphi_{\mathrm{p}i})}$
0–0.4	21	0.08	19.60	0.10
0.4–0.8	71	0.22	53.08	6.05
0.8–1.0	39	0.14	35.01	0.45
1.0–1.2	41	0.14	35.15	0.99
1.2–2.0	61	0.32	79.28	4.21
>2	12	0.03	7.35	2.94
			Statistics: $\chi^2 = \sum_i \chi_i^2 = 14.74$	
Distribution fractile $\chi^2_{m,1-\alpha}$; $\alpha = 0.005$, $m = 4$: 14.86				

in the ensemble corresponds to $F(0) = 0.024$. According to Eq. (3) the theoretically calculated probability of occurrence of zero concentration values is $F(0) = 0.019$. Consequently, there are sufficiently strong reasons to give preference to the continual law of distribution taking into account the effect of the concentration intermittence when analyzing the data on total protein concentration.

The probability of observing $k = 0$ microorganisms depends on the sample volume and makes up $p(0) = \exp(-\overline{k})$. In our case, the typical culturable microorganism concentration is $\log_{10}(C_{\mathrm{b}}) \approx 3.7$, which corresponds to $\overline{k} \approx 5000$ microorganisms per $1\,\mathrm{m}^3$. The volume of collected air samples is about $0.75\,\mathrm{m}^3$. The estimates show that probability of observing a zero number of culturable microorganisms in our samples should be negligible. At the same time, a $\frac{1}{50}-\frac{1}{250}$ part of the sample is used when seeding each sample to a nutrient medium. Therefore, the probability of detecting on a medium of $k=0$ microorganisms is approximately 3×10^{-7}. This probability is also very small. However, in our samples a zero number of culturable microorganisms is observed in some cases. We think that the question concerning the presence of the effect of intermittence of the concentration of culturable microorganisms aerosols should be further investigated.

Thus, the hypothesis about different physical natures of the statistics of total protein concentration and culturable microorganism concentration is confirmed. The statistics of culturable microorganism concentration is described with a discrete law—Poisson distribution. The statistics of total protein concentration obeys the laws of continual statistics.

3. Correlation characteristics of bioaerosol concentration fields

Table 5 presents correlation coefficients of total atmospheric protein concentration and the decimal logarithm of culturable microorganism concentration. It is seen that correlation coefficients of total protein concentration and correlation coefficients of the decimal logarithm of culturable microorganism concentration are always positive, and no obvious dependence on the flight altitude is revealed. Correlation coefficients of the concentration of total protein aerosols are in the range of 0.41–0.87. Correlation coefficients of the concentrations of culturable microorganism aerosols are in the range of 0.18–0.74. This may indicate that sources of bioaerosols are remote ones and they quite fully mix as particles wander in troposphere.

Table 5
Coefficients of correlation of total protein concentration and correlation coefficients of decimal logarithm of culturable microorganisms concentration calculated for time series of data[a]

Altitude (km)	Altitude (km)							
	0.5	1	1.5	2	3	4	5.5	7
0.5	1	0.65	0.70	0.57	0.50	0.61	0.55	0.53
1	0.59	1	0.55	0.60	0.50	0.54	0.56	0.57
1.5	0.19	0.27	1	0.56	0.54	0.57	0.57	0.41
2	0.49	0.56	0.45	1	0.75	0.76	0.80	0.65
3	0.48	0.54	0.26	0.74	1	0.87	0.64	0.62
4	0.27	0.54	0.34	0.33	0.54	1	0.77	0.68
5.5	0.51	0.50	0.37	0.67	0.51	0.39	1	0.73
7	0.55	0.50	0.18	0.47	0.65	0.39	0.47	1

[a]Data shown in gray correspond to correlation coefficient of microorganism concentration.

Table 6
Coefficients of cross correlation of total protein concentration and decimal logarithm of culturable microorganisms concentration calculated for time series of data

Altitude (km)	Altitude (km)							
	05	1	15	2	3	4	55	7
0.5	0.18	0.10	0.39	0.23	0.20	−0.01	0.23	0.33
1	0.18	0.05	0.06	0.21	0.13	−0.17	0.14	0.19
1.5	0.24	0.19	0.21	0.33	0.40	0.03	0.33	0.25
2	0.30	0.12	−0.07	0.39	0.37	−0.07	0.32	0.39
3	0.18	0.14	0.07	0.41	0.37	0.01	0.35	0.25
4	0.24	0.18	0.17	0.51	0.36	−0.05	0.36	0.26
5.5	0.34	0.13	−0.03	0.50	0.39	−0.05	0.32	0.31
7	0.09	0.03	0.06	0.35	0.23	−0.08	0.13	0.14

Table 6 presents the coefficients of cross correlation of total protein concentration and decimal logarithm of culturable microorganism concentration calculated for time series of data. It is seen that though there are 13% of negative values, positive coefficients of cross-correlation prevail. The value range of cross-correlation coefficients indicates weak statistical dependence of concentration fields of total protein and culturable microorganism aerosols. Thus, it can be supposed that particles containing atmospheric protein and aerosol particles containing culturable microorganisms originate from different sources.

4. Wavelet and harmonic analyses of bioaerosol concentration fields

Wavelet transformation of random sequences provides a two-dimensional evolution of the studied one-dimensional signal; the scale and the coordinate are considered as independent variables. As a result, there appears the possibility of analyzing the signal characteristics in the time and the frequency spaces simultaneously (Torrence & Compo, 1998).

Wavelet transformation $Wf(x,a)$ of a one-dimensional signal $f(t)$ consists in its basis expansion constructed from a special function (wavelet)—Ψ by scale changes a and transfers x

$$Wf(x,a) = \frac{1}{a}\int_{-\infty}^{+\infty} \Psi\left(\frac{t-x}{a}\right) f(t)\,\mathrm{d}t. \tag{6}$$

Each of the functions of this basis characterizes a certain time scale a and its localization in the physical space (time) x. Unlike Fourier transformation, wavelet transformation provides for two-dimensional evolvement of the analyzed one-dimensional signal; the scale and coordinate are considered as independent variables. This allows us to analyze the signal characteristics simultaneously in the time and the frequency spaces (Torrence & Compo, 1998). We used the Morle wavelet to analyze variations of the concentration:

$$\Psi(t) = \frac{1}{2\pi}\exp\left(\mathrm{i}\omega_0 t - \frac{t^2}{2}\right). \tag{7}$$

In the general case, the choice of the analyzing wavelet is ambiguous and depends on the type of task. We chose the Morle wavelet due to the following reasons:

1. It is good for analysis of quasi-periodic processes as it has good localization in the frequency space;
2. The mother function is a periodic signal modulated by the Gaussian function, that is why we can compare the wavelet spectrum with the spectrum of atmospheric waves, which are own oscillations atmosphere (Monin, 1969) usually considered to be quasi-periodic;
3. Correct choice of its parameters enables us to avoid complex recalculations of the time-scale during the process, which allows us to use the time scale as a specified value in carrying out harmonic analysis; the presence of a complex component allows us to analyze not only the amplitudes but also the phases of the isolated harmonics;
4. And, finally, we have already successfully used it to analyze the near-ground concentration fields of atmospheric aerosols (Borodulin, Safatov, Khutorova, Kutzenogii, & Makarov, 2003a).

The Morle wavelet also provides pictorial presentation and easy interpretation of the data in the terms of Fourier analysis. In the works (Khutorova & Teptin, 2001; Khutorova & Korchagin, 2001) wavelet analysis of long series of measurements of aerosol and chemical pollutants by the data of atmospheric monitoring stations revealed great potential for investigating time variations of atmospheric pollutant concentrations. The analysis of the data of the on-land concentration of total protein aerosols obtained in Novosibirsk environs and presented in the work (Borodulin et al., 2003a) allowed us to reveal a relation between the variations of mass concentration of aerosols and total protein concentration in the on-land atmospheric layer with its typical periodic synoptic processes.

In addition to the wavelet analysis, we also performed the harmonic analysis of the time series to evaluate the amplitudes and phases of their periodic components (Jenkins & Watts, 1971). In this case, the time series is approximated by the sum of harmonics with already known periods. Usually, in such analyses the periods are from the suppositions of the selected model of atmospheric physics or are found with other methods. In the general case, we can perform the wavelet analysis of the time series, determine

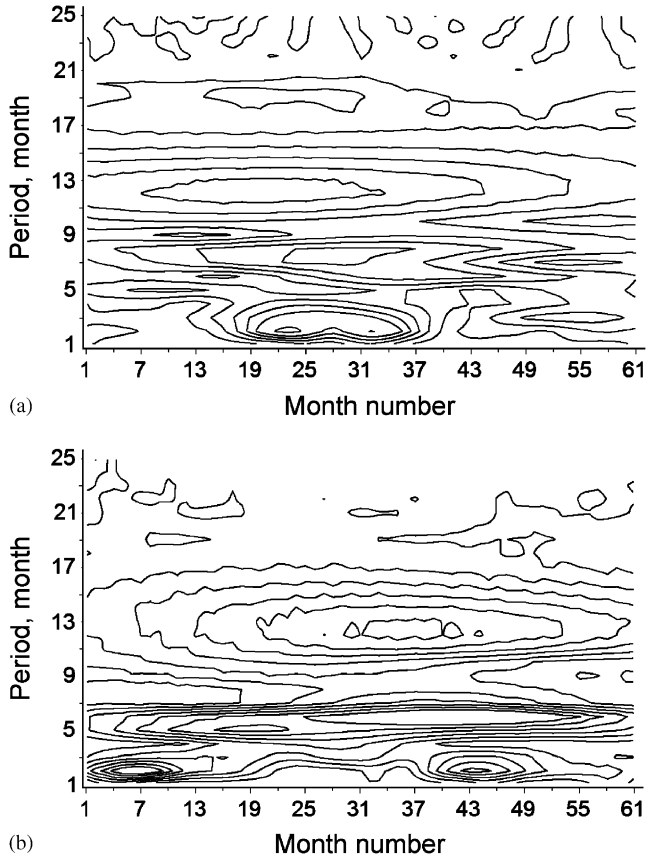

Fig. 3. Wavelet transformation of time series of concentrations of total protein and live microorganism aerosols for the altitude of 7 km (a) and (b), respectively.

the existence of stable oscillations and their time scales with this method, and then find the amplitudes and phases of periodic variations by means of harmonic analysis. The method of harmonic analysis was successfully used, for example, in (Khutorova & Korchagin, 2001) to find the parameters of tidal processes and seasonal variations of wind velocity fields in the lower thermosphere.

In our case, the time series was also approximated with the sum of harmonics with P_i obtained as a result of the wavelet analysis

$$S(t) = A_0 + \sum_i A_i \cos\left[(t - T_i)\,\frac{2\pi}{P_i}\right], \tag{8}$$

where A_0 is the constant component of the signal, A_i and T_i are amplitudes and phases of harmonics found with the least-squares method. The periods are usually preset from the suppositions of the selected model of physics of atmosphere or are found with other methods.

The relation between the scale of wavelet transformation a with the Morle wavelet and the corresponding Fourier wavelength λ is expressed with the formula (Torrence & Compo, 1998)

$$\lambda = \frac{4\pi a}{\omega_0 + \sqrt{2 + \omega_0^2}}, \tag{9}$$

where ω_0 is the parameter of the maternal function from Eq. (7). The module of wavelet transformation characterizes the time variation of the relative contribution of components of different scales to the studied signal, i.e. at each moment we can estimate the intensity of variations of all the analyzed time scales a.

Thus, wavelet analysis of a time series reveals the existence of stable fluctuations and their time scales, and harmonic analysis allows the amplitudes and phases of the revealed periodic processes to be determined.

Fig. 3 presents an example of wavelet spectra obtained for total protein and culturable microorganism concentrations for the year 2000 at the altitude of 7 km. The wavelet spectrum for a number of total protein concentrations was constructed for the range of its variation from 0.1 to 1.1 with a step of 0.15. Wavelet spectrum for a number of culturable microorganism concentrations was constructed for the range of 0.05–0.35 with a step of 0.05. The analysis of the obtained data shows that the wavelet analysis for each time series reveals regular periodic seasonal processes: 12, 8.5, 6 and 4 months. The amplitude of these variations is not constant in time. Besides regular fluctuations, sometimes there are processes with a period of about 24 months, i.e. inter-year fluctuations. We note the approximate similarity of the obtained wavelet spectra of total protein and culturable microorganism concentrations. This presupposes that variations in the bioaerosols mass concentration are largely determined by dynamic processes in troposphere, in particular by seasonal fluctuations. At the same time, incomplete similarity between wavelet spectra may be indicative of different natures of sources of tropospheric bioaerosols.

When carrying out the harmonic analysis of concentration series, experimental data were approximated with the sum of seasonal harmonics with periods of 12, 8.5, 6 and 4 months. It means that we took the harmonics that were significant in wavelet spectra for all time series. Fig. 4 shows an example of approximation of the time series of total protein and culturable microorganism concentrations with the sum of harmonics for the year 2000 at the altitude of 0.5 km. Blanks in Fig. 2 indicate the absence of data. Tables 7 and 8 give the summary results of harmonic analysis: A_0 is the constant component and confidence interval; A_i and T_i are amplitudes and times of the maximum and confidence intervals for the periods of 12, 8.5, 6 and 4 months, respectively, see the figures at the top and the bottom of the tables; the residual portion of the total distribution (percentage) without taking into account the seasonal variations and the constant component is presented in the last line. The data analysis shows that seasonal variations cause approximately 80% of the total distribution of variations of the total protein concentration. The variation in amplitudes of culturable microorganism concentration are small as compared to the constant component for all altitudes. The harmonic analysis shows that not all amplitudes of seasonal fluctuations are steady during the observation period. The evaluations showed that all the harmonics except for the 4-month ones were surface ones. It means that fluctuations are in phase at the given observation altitudes. It was revealed that the 4-month periodicity was internal and was caused by a wave spreading from above with a vertical rate of about 4 km/month.

Thus, like in (Borodulin et al., 2003a), the wavelet and harmonic analyses of 5-year time series of tropospheric bioaerosol concentration showed at the qualitative level that variations of total protein and

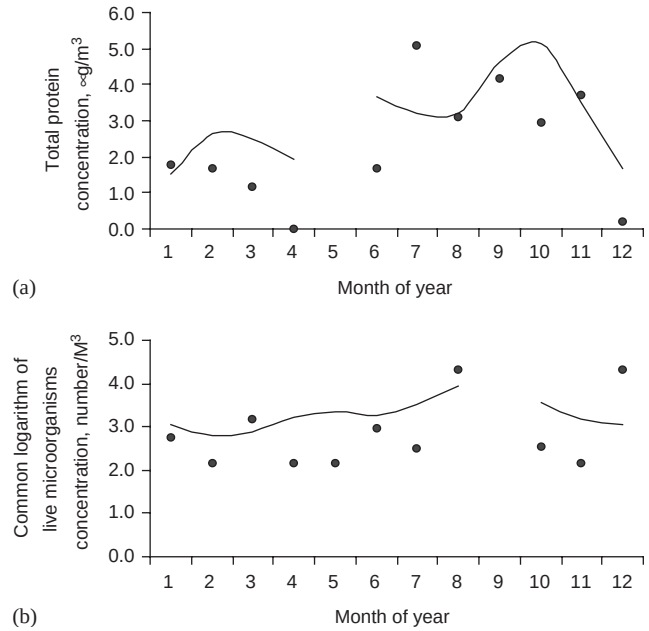

Fig. 4. Approximation of experimental series of concentrations of total protein and live microorganism aerosols with the sum of harmonics with revealed seasonal periods for the altitude 0.5 km (a) and (b), respectively.

culturable microorganism concentrations in the South of Western Siberia are mainly determined by the revealed seasonal periodic processes.

5. Conclusion

The analysis of the data on concentration fields of tropospheric bioaerosols shows that the concentration of total protein aerosols obeys the laws of continual statistics, and the concentration of culturable microorganism aerosols obeys the laws of discrete statistics. The performed analysis of correlation coefficients of bioaerosol concentration fields confirms our previous hypothesis that sources of bioaerosol are remote, and they quite fully mix as particles wander in troposphere. The value range of cross-correlation coefficients of total protein and culturable microorganisms aerosols shows that statistical relation between these values is weak. This can be explained by the fact that particles containing atmospheric protein and aerosol particles containing culturable microorganisms may originate from different sources. Wavelet analysis of the data showed that variations of bioaerosol concentration in the troposphere are mainly caused by typical seasonal processes with the periods of 12, 6, 4 and 8–9 months. Seasonal variations cause approximately 80% of the total distribution of variations of total protein concentration, and the variation amplitudes of culturable microorganism concentration are small as compared to the constant component. The performed harmonic analysis of bioaerosol concentration fields revealed that the 4-month periodicity was internal and was caused by a wave spreading from above with a vertical rate of about 4 km/month.

Table 7
Summary results of harmonic analysis of series of total protein concentrations

Altitude (km)	0.5	1	1.5	2	3	4	5.5	7
A_0	2.83	2.5	2.13	2.09	1.92	1.79	2.07	1.85
	6.97×10^{-10}	1.43×10^{-10}	3.90×10^{-10}	6.47×10^{-10}	5.55×10^{-10}	5.39×10^{-10}	4.55×10^{-10}	4.93×10^{-10}
A_1	1.43	1.47	1.14	1.27	1.53	1.3	1.28	0.93
	1.58×10^{-9}	1.20×10^{-9}	8.52×10^{-10}	5.63×10^{-10}	7.60×10^{-10}	1.24×10^{-10}	6.30×10^{-10}	3.18×10^{-10}
T_1	240.79	228.01	244.21	266.37	265.03	264.8	241.43	261.56
	6.57×10^{-8}	4.33×10^{-8}	3.77×10^{-8}	3.41×10^{-8}	5.41×10^{-8}	8.57×10^{-9}	2.80×10^{-8}	1.98×10^{-8}
A_2	0.57	0.57	0.36	0.73	0.79	0.67	0.9	0.52
	8.46×10^{-10}	4.06×10^{-10}	1.22×10^{-10}	7.36×10^{-10}	8.73×10^{-10}	6.30×10^{-10}	7.59×10^{-10}	3.96×10^{-10}
T_2	176.31	161.28	194.42	175.61	195.95	201.8	170.74	171.42
	3.23×10^{-8}	1.04×10^{-8}	1.84×10^{-8}	2.29×10^{-8}	9.48×10^{-8}	7.88×10^{-8}	1.65×10^{-8}	1.30×10^{-8}
A_3	0.39	0.26	0.37	0.43	0.42	0.42	0.47	0.31
	9.26×10^{-10}	9.63×10^{-10}	8.16×10^{-10}	7.83×10^{-10}	6.77×10^{-10}	6.18×10^{-10}	8.07×10^{-10}	5.49×10^{-10}
T_3	118.58	160.31	110.4	87.72	102.32	117.82	28.61	36.58
	1.93×10^{-8}	2.43×10^{-8}	1.36×10^{-8}	5.65×10^{-9}	1.26×10^{-8}	1.12×10^{-8}	3.60×10^{-9}	3.71×10^{-9}
A_4	0.81	0.72	0.61	0.62	0.46	0.36	0.16	0.3
	3.84×10^{-10}	9.45×10^{-10}	3.23×10^{-10}	7.54×10^{-10}	5.36×10^{-10}	2.47×10^{-10}	4.16×10^{-10}	5.58×10^{-10}
T_4	64.34	74.03	57.26	68.88	68.27	55.12	116.23	84.23
	7.00×10^{-9}	1.07×10^{-9}	6.00×10^{-9}	7.08×10^{-9}	6.69×10^{-9}	5.07×10^{-9}	4.34×10^{-9}	9.21×10^{-9}
Residual dispersion (%)	15.86	21.81	17.6	23.35	34.64	28.16	15.56	18.48

Table 8
Summary results of harmonic analysis of series of culturable microorganism concentrations

Altitude (km)	0.5	1	1.5	2	3	4	5.5	7
A_0	3.3	3.36	3.19	3.18	3.17	3.21	3.36	3.21
	4.79×10^{-10}	4.36×10^{-10}	4.31×10^{-10}	4.91×10^{-10}	4.70×10^{-10}	4.65×10^{-10}	4.39×10^{-10}	4.35×10^{-10}
A_1	0.4	0.4	0.32	0.48	0.28	0.11	0.52	0.45
	3.88×10^{-10}	3.60×10^{-10}	3.76×10^{-10}	4.11×10^{-10}	1.4×10^{-10}	3.28×10^{-10}	4.06×10^{-10}	2.89×10^{-10}
T_1	234.7	218.82	197.37	231.73	267.28	358.05	227.36	233.02
	1.53×10^{-8}	1.06×10^{-8}	7.46×10^{-8}	1.61×10^{-8}	1.55×10^{-8}	1.30×10^{-8}	1.45×10^{-8}	1.18×10^{-8}
A_2	0.18	0.15	0.24	9.69×10^{-3}	0.2	0.18	0.14	0.23
	3.60×10^{-10}	5.45×10^{-10}	3.70×10^{-10}	3.90×10^{-10}	4.57×10^{-10}	4.61×10^{-10}	3.67×10^{-10}	2.57×10^{-10}
T_2	126.1	32.5	131.23	142.28	167.41	113.67	225.88	136.35
	3.78×10^{-9}	2.82×10^{-9}	6.97×10^{-9}	8.94×10^{-9}	1.22×10^{-8}	7.80×10^{-9}	1.32×10^{-8}	4.98×10^{-9}
A_3	0.12	0.16	0.08	0.17	0.16	0.11	0.05	0.5
	3.00×10^{-10}	2.76×10^{-10}	1.14×10^{-10}	2.35×10^{-10}	2.95×10^{-10}	1.17×10^{-10}	2.53×10^{-10}	3.79×10^{-10}
T_3	123.03	121.23	175.1	9.23	121.39	121.42	156.82	86.6
	7.43×10^{-9}	6.31×10^{-9}	5.63×10^{-9}	3.97×10^{-9}	5.71×10^{-9}	3.20×10^{-9}	6.35×10^{-9}	4.41×10^{-9}
A_4	0.17	0.11	0.04	0.08	0.16	0.03	0.25	0.15
	4.43×10^{-10}	2.49×10^{-10}	3.34×10^{-10}	2.45×10^{-10}	1.47×10^{-10}	3.01×10^{-10}	2.62×10^{-10}	3.22×10^{-10}
T_4	16.8	89.52	41.74	117.9	34.67	115.07	108.94	112.56
	1.19×10^{-9}	2.31×10^{-9}	2.47×10^{-9}	2.00×10^{-9}	1.27×10^{-9}	6.16×10^{-9}	4.17×10^{-9}	6.32×10^{-9}
Residual dispersion (%)	17.19	9.05	12.59	14.76	11.43	4.08	17.7	29.12

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