## Vertical profiles of bioaerosol concentration in the troposphere over the south of Western Siberia

A.I. Borodulin,<sup>1</sup> A.S. Safatov,<sup>1</sup> B.D. Belan,<sup>2</sup> M.V. Panchenko,<sup>2</sup> V.V. Penenko,<sup>3</sup> and E.A. Tsvetova<sup>3</sup>

<sup>1</sup>Research & Development Institute of Aerobiology,

State Scientific Center of Virology and Biotechnology "Vector," Kol'tsovo, Novosibirsk Region <sup>2</sup> Institute of Atmospheric Optics,

Siberian Branch of the Russian Academy of Sciences, Tomsk <sup>3</sup> Institute of Computational Mathematics and Mathematical Geophysics, Siberian Branch of the Russian Academy of Sciences, Novosibirsk

Received February 9, 2005

Analysis of the vertical profiles of the concentrations of aerosols of total protein and viable microorganisms is presented. The results were obtained by averaging the data array on the concentration of atmospheric bioaerosols collected by the airplane probing of the troposphere over the southwest of Siberia in 1999-2003. It is shown that the vertical profiles of bioaerosol concentration present a unique footprints of the state of the atmosphere over the northern hemisphere, which reflect the information about the sources of bioaerosols and the processes of their transformation in the atmosphere.

## Introduction

Research & Development Institute of Aerobiology at the State Scientific Center "Vector" together with the Institute of Atmospheric Optics SB RAS carry out systematic monitoring of tropospheric bioaerosols over the south of Western Siberia since 1998. Collection of atmospheric air samples is carried out in the end of every month at the point with the coordinates 54°23'N and 82°09'E at the heights of h = 0.5; 1; 1.5; 2; 3; 4; 5.5, and 7 km from onboard AN-30 "Optik-E" aircraft-laboratory. Then the contents of total protein and viable microorganisms in them are determined. The problems of the generalized technique used and results of investigations have been earlier presented in Refs. 1 and 2.

The obtained data array enabled us to reveal seasonal variations of the bioaerosol concentration in the troposphere. In particular, it was shown that the total amount of atmospheric protein has minimum in winter. In spring, when animals and vegetation awaken, it monotonically increases and reaches maximum, which is an order of magnitude greater than the winter values, in summer or in the beginning of fall season. Late in the fall, the atmospheric protein concentration decreases to the level. The initial monthly mean protein concentrations smoothly (during 4–5 months) increase up to the maximum value observed in August-September, and then dramatically (during 2 months) decrease to the typical winter values. This does not agree with the maximum activity of animals and vegetation in the region, which is observed since May and until August. The prevalence of winds from southwest in our region makes the hypothesis about

essential effect of the atmospheric protein sources located out of the territory of Western Siberia quite realistic. Indeed, if follow the wind rose, such sources should be situated in the region of Central Asia, where animals and vegetation have maximum activity during the most part of year. Analogous conclusions were drawn for aerosols of viable microorganisms.

For quantitative justification of these suppositions, a cycle of investigations on estimation of the possible location of the corresponding sources was carried out by means of the methods of inverse simulation in combination with the methods of theory of sensitivity of models and functionals. Investigations were carried out based on the complex of mathematical models developed at ICMMG SB RAS. The semi-spherical models of admixture transport in the atmosphere constructed in the frameworks of Euler and Lagrange approaches were used.<sup>3,4</sup>

One can select three groups among the methods mathematical simulation used in solving the of problems of the "receptor-source" type:

1) the back trajectories methods;

2) the methods based on the use of conjugated equations in Marchuk's interpretation;

3) the methods based on variational principle, which generate combined algorithms for direct and inverse modeling, as well as the methods of the theory of sensitivity.

The traditional method for simulation of the trajectories in the frameworks of the Lagrange approach is quite well described (see, for example, the reviews<sup>5-7</sup> and references therein). Most often, the trajectories of particles emitted from a source are calculated by this technique. Application of this method to calculation of back trajectories is restricted by the fact that the notion of trajectories in reverse time at solving the problems of the "receptor-source" type is not correct for the models of admixture transport and turbulent exchange. It makes sense to speak about back trajectories based on their traditional concept. Those are calculated from the velocity vector in reverse time, only for the advective part of the models of admixture transport and assuming that the carrier medium is incompressible. Let us note that it is taken into account in the HYSPLIT modeling system,<sup>8,9</sup> which is widely used by users because it is free for access on the NOAA website. The problem of trajectories was discussed in a more detail in Ref. 10, where the example was presented confirming the thesis, that the trajectory methods are characterized by extremely high sensitivity to variations of the input parameters. As the author noted, randomization of the trajectories occurs even at modeling in direct time on determinate spatially smooth velocity fields. This fact casts some doubts on the applicability of the method of back trajectories to the problems of "receptor-source" type.

From this standpoint, the second direction has an advantage, because it works with the models of Euler type.<sup>10–12</sup> The main element in it is the conjugated problems determined by the duality relationships for linear functional of the type of the Lagrange identity, which are solved in reverse time. The functions are usually set as the sources in conjugated problems, which represent the configuration and information concept of the receptors.

The approach developed at ICMMG SB RAS<sup>3,4,13,14</sup> is more versatile. It is based on variational principle in combination with the methods of sensitivity and it is applicable to linear and nonlinear functionals and the models of both Euler and Lagrange types. Let us note that in this approach the models of the processes are considered as restrictions to the set of functions of states and parameters. In its turn, conjugated functionals play the role of generalized Lagrange factors for taking into account different restrictions in the frameworks of the variational principle. The "models of observations," which present the relationships between the values and functions measured in the receptor-areas and the functions of states of the models of the processes, also take part in forming the functionals.

The core of this approach is the combined algorithm for direct and inverse simulation.<sup>13</sup> The conjugated problems in it are generated by the variational principle for the selected set of functionals, and the functions of sensitivity are calculated using the solutions of appearing direct and conjugated problems. One of modifications of the general algorithm destined for finding of the sources is described in Refs. 14 and 15.

The original method for inverse simulation in Lagrange statement based on the variational principle and solution of the generated conjugated problems<sup>3,4</sup> is applied in this paper for computational experiments

on tracing the remote transfer of admixtures. The algorithms with implicit scheme of second order of accuracy in time for determinate calculation of trajectories in advective-transfer part of the model of processes and the algorithm of the Monte Carlo type for turbulent part of the model are combined in it. This complex is constructed on the principles of local approximation of the direct and conjugated equations in approximation of the method of splitting the model as a whole. The detailed description of the method can be found in Refs. 3 and 4.

Summing up the analysis of peculiarities of different methods, it is worthy to emphasize once more that one can work with advective-diffuse models in reverse time only using conjugated problems. These are just the algorithms (theoretically and structurally), which provide for specific data on the relations between the receptors and sources. To organize the scenarios of simulation, the results measured at individual heights and total over the height and time of carrying out the observation experiments were presented in the form of functionals on the set of functions of state. The scenarios of inverse simulation were calculated to 30 days back in time from the moment of making the observations.

The spatiotemporal structure of the atmosphere was reconstructed with the step of 30 minutes by means of the system<sup>14</sup> based on joint use of the model of hydrothermodynamics of the atmosphere in the coordinate system following the relief of the Earth's surface and the data of NCEP/NCAR<sup>16</sup> Reanalysis for these years. The functions of sensitivity for the selected set of functionals to variations of the source parameters were calculated by means of the models of Euler type in the mode of inverse simulation. Then, the areas of possible arrangement of the sources of admixture, from which the observed concentrations of bioaerosols could be recorded, were estimated using the levels of significance and the carrier-areas of these functions.

The conjugated (in reverse time) trajectories, originated from the points of measurements where the samples had been collected, were calculated by the determinate-stochastic models of Lagrange type. These models are the combinations of the algorithms of determinate transfer of substances along the trajectories of motion of air masses and the Monte Carlo method representing the stochastic structure of turbulence.

We had in mind the following ideas in selecting the duration of 30 days in our numerical experiments:

1) the estimates of the characteristic lifetimes of the substances under study;

2) the calculated results on the functions of sensitivity.

The levels of significance of these functions were analyzed in different variants of setting the functionals for description of the receptors and the regime of observations. As a result, a trade-off variant was selected.

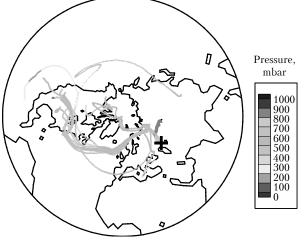
The step of calculations of 30 minutes was determined as the basis. It is acceptable according to

the conditions of approximation. However, it should be noted that all algorithms in our complex of models used for calculations are adaptive. So the time steps in the split schemes were selected depending on the intensity of the processes. The interval of 30 minutes was set for meeting all stages of the split operating with "under-grid," relative to the basis interval, time steps. Such an organization of the algorithms was dictated by the variational principle.

Analysis of the behavior of the calculated trajectories shows that air masses coming to the sample collection point have different prehistory. During the motion, they are situated at different heights, over different parts of the surface (often even over different continents and oceans), and only approaching the point of sampling the trajectories approach each other and come together to the same point. Hence, aerosol particles emitted to the atmosphere from different sources really can come to the point of observation. As particles of the micrometer size can reside in the atmosphere during quite a long time, they can reach the point of observations together with air masses.

During the transportation process, the particles move from one height to another, come to the nearground layer (where, obviously, they are enriched with bioaerosols), their intense mixing (these places are different for each back trajectory) occurs, then such masses reach the point of measurement. The example of conjugated back trajectories is shown in Fig. 1, in which it is seen that aerosols generated not only over Middle Asia, but over Africa and even over America can come to the region of observations. It should be noted that one has to consider the calculated result as a tentative estimate of the possible routes of migration of admixtures.

Conjugated problem, Lagrange's variant of the transportation model Four-dimensional back trajectories, 1–29 of July, 1999



54.23°N, 82.09°E, 1500 m, July 29, 1999, 08:00 of Greenwich time.

Fig. 1. An example of back trajectories of the air mass transport calculated for the height of 1.5 km for July 29, 1999. The cross in the figure shows the point of sampling.

To find the specific sources, which are responsible for appearance of the substances identified in observations, it is insufficient to have only the data acquired with the receptor or a set of close receptors, as it occurs in the considered series of observation experiments. In order to increase reliability of the conclusions to be drawn, it is necessary to plan the observation experiments and to have a possibility of carrying out measurements using additional receptors.<sup>10,14</sup>

Qualitative analysis of the obtained data has shown that the values of the total protein concentration have significant scatter, and no explicit dependence of the bioaerosol concentration on the height of observations is observed in some experiments. The observed scatter of the values is caused by the statistical nature of the process of spread of atmospheric admixtures, the atmospheric turbulence, and the errors in measurements leading to additional scatter of the observed values of concentration.

The statistical properties of the bioaerosol concentration fields were studied in Ref. 17. In particular, it was shown that the distribution of the total protein aerosol concentration obeys the laws of continuous statistics: it is described by the analytical solutions of the Fokker-Plank-Kolmogorov equation<sup>18</sup> and can be sufficiently accurate approximated by the logarithmic-normal law. In its turn, the distribution of the viable microorganism aerosol concentration is adequately described in the frameworks of the discrete Poisson statistics.

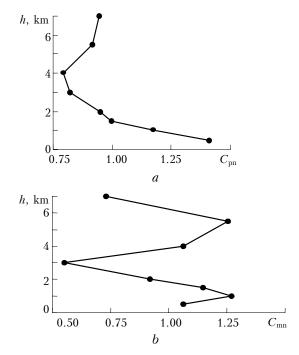
The standard deviation of the errors in measurements of the total protein aerosol concentration is 0.2  $\mu$ g/m<sup>3</sup> from the observed values. The errors in determination of the viable microorganism concentration are traditionally estimated by the 95% confidence interval, within which the scatter of decimal logarithm of the observed concentration values measured in  $1/m^3$  lies. In our case the order of magnitude of the 95% confidence interval is less than a half of the decimal logarithm.

The vertical profiles of the total protein and viable microorganism aerosol concentration are presented in this paper, and their properties are discussed. The profiles were obtained by averaging of the array of observational data on the tropospheric bioaerosol concentration collected in 1999–2003.

The bioaerosol sources are situated in quite a thin near-surface layer of the atmosphere, and are mainly caused by activity of animals and vegetation. If there were no effect of circulation processes in the atmosphere, the profiles of the observed bioaerosol concentration would steadily decrease with height.

The normalized vertical profiles of the total protein aerosol concentration  $C_{\rm pn}$  and the viable microorganism aerosol concentration  $C_{\rm mn}$  are shown in Fig. 2. The data were obtained by averaging over the years of measurements with subsequent normalization of the monthly mean data to the mean

values over the heights, and then they were finally averaged over the months of observations. Normalizing to the mean values over heights was carried out for excluding the effect of seasonal variations of the concentration on the vertical profiles. Averaging over years and months enables one to reveal some features of the vertical variations of the bioaerosol concentration, which are not observed in individual realizations.



**Fig. 2.** Normalized vertical profiles of the total protein aerosol concentration,  $C_{pn}$ , and the concentration of aerosol comprising the viable microorganisms,  $C_{mn}$ .

It follows from Fig. 2a that the 5-year total profile of the total protein aerosol concentration has maximum at the height of 0.5 km, and then the concentration decreases with height. It is an evidence of the fact that the main sources of the total protein aerosols are really situated near the underlying surface. However, the sources of the total protein aerosols work both in our region and in the regions far from the point of observations. This is confirmed by significant amount of the total protein in winter, when the ground in Western Siberia is covered with snow.

The normalized vertical profiles of the total protein aerosol concentration  $C_{\rm pn}$  and the viable microorganism aerosol concentration  $C_{\rm mn}$  are shown in Fig. 3 separately for each year of measurements. For convenience of data analysis, the profiles of each next year are displaced to the right from the data of 1999.

This property manifests itself to a certain degree in the annual vertical profiles of the concentration (Fig. 3*a*). It is seen in Figs. 2a and 3a that some local maxima of concentration caused by complicated trajectories of the air mass motion in the atmosphere (see discussion in introduction to the paper) are superposed to both the total profile for 1999–2003 and the annual profiles. Let us also note that practically monotonic decrease of the concentration with height was observed in 2001.

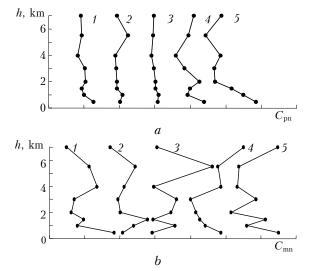


Fig. 3. Normalized vertical profiles of the concentrations of total protein aerosol  $C_{pn}$  (*a*) and viable microorganisms  $C_{mn}$  (*b*) as functions of the year of observation. Numbers 1-5 mark years 1999–2003.

In their turn, the vertical profiles of the viable microorganism aerosol concentration (see Figs. 2b and 3b) show that the regularities of their field formation, possibly, are more complicated in comparison with the total protein aerosols (Figs. 2a and 3a). The tendency toward the decrease of the viable microorganism aerosol concentration with height is practically absent in the vertical profiles of  $C_{\rm mn}$ . At the same time, they have maxima of concentration caused by contributions from numerous "elevated effective" sources. There are two effective heights of the sources in the 5-year total profile  $C_{\rm mn}$ : 1.0 and 5.5 km.

Starting from 2001, we also have carried out monitoring of the atmospheric bioaerosols on the territory of SSC VB "Vector" in Kol'tsovo village, Novosibirsk Region. In spite of the fact that monthly measurements on the "Vector" territory, different from airborne sounding, are carried out in the middle of month, the relief is different and the measurement points are about 100 km far from each other, it is interesting to compare the annual average values of the bioaerosol concentration in order to add the data point to the vertical profiles of the concentration.

The mean values and standard deviations of the total protein aerosol concentration and decimal logarithm of the viable microorganism concentration are given in Table 1. They were obtained in the near-ground layer on the territory of SSC VB "Vector" and at the point of airborne sounding at the height of 0.5 km were averaged over 2001, 2002, and 2001–2003.

	Concentration			
Year	Total protein, $\mu g/m^3$		Viable microorganisms, decimal logarithm, $1/m^3$	
	Near-ground layer	$h = 0.5  {\rm km}$	Near-ground layer	h = 0.5  km
2001	$2.07 \pm 2.84$	$3.41\pm3.34$	3.32±3.14	3.38±1.10
2002	0.38±0.36	2.23±2.18	$3.52\pm0.69$	$3.47 \pm 0.94$
2003	0.32±0.33	$3.38 \pm 2.50$	3.27±0.63	3.37±0.61
2001-2003	1.36±2.18	$3.01 \pm 2.72$	3.15±0.62	$3.34\pm0.86$

Table 1. Mean values plus-minus standard deviation of the bioaerosol concentrationin the near-ground layer on the territory of SSC VB "Vector" and at the point of airbornesounding at the height of 0.5 km averaged over the years of measurement

It was noted above that if one ignores the complicated dynamics of air mass mixing, the values of concentration in the near-ground layer of the atmosphere should be greater than that at the height of 0.5 km. It is seen from the data presented in Table 1 that the inverse dependence is observed for the total protein aerosols, but the data on the viable microorganism aerosol concentration obtained in 2002-2003 and averaged over 2001-2003 at these levels are comparable in the magnitude. Hence, comparison of the data on the concentrations of the total protein and viable microorganisms in the nearground layer and at the lower boundary of the troposphere is an evidence of essential effect of the atmospheric processes with the resulting determining role of remote sources in the formation of the bioaerosol background in our region.

Thus, each of the vertical profiles of the bioaerosol concentration obtained, including annual ones, is a unique portrait of the state of the atmosphere of the Northern hemisphere, which contains the generalized information about numerous possible bioaerosol sources and about the processes of their transformation in the atmosphere.

## Acknowledgments

The work was supported in part by the Russian Foundation for Basic Research (grants No. 04–05– 64562, 04–05–64559, 04–05–08010), the Program of Presidium SB RAS No. 13 and Department of Mathematics RAS No. 1.3.2; interdisciplinary project SB RAS No. 130 "Ecological problems of Siberian cities." Authors would like to thank the aforementioned foundations for the support of this work.

## References

1. I.S. Andreeva, B.D. Belan, A.I. Borodulin, G.A. Buryak, Yu.V. Marchenko, S.E. Ol'kin, M.V. Panchenko, V.A. Petrishchenko, O.V. P'yankov, I.K. Reznikova, A.S. Safatov, A.N. Sergeev, and E.V. Stepanova, Atmos. Oceanic Opt. **13**, Nos. 6–7, 592–596 (2000).

2. I.S. Andreeva and A.I. Borodulin, Khimiya v Interesakh Ustoichivogo Razvitiya **10**, No. 5, 547–561 (2002).

3. V.V. Penenko and E.A. Tsvetova, Atmos. Oceanic Opt. **12**, No. 6, 462–468 (1999).

4. V.V. Penenko and E.A. Tsvetova, Prikl. Mekh. Tekh. Fiz. 41, No. 5, 161–170 (2000).

5. R.A. Pielke, W.A. Lyons, R.T. McNider, M.D. Moran, D.A. Moon, R.A. Stocker, R.L. Walko, and M. Uliasz, in: *Proc. NATO/CCMS International Technical Meeting on Air Pollution Dispersion Modeling and Its Application* (British Columbia, Vancouver, Canada, 1990), pp. 1–33.

6. H.C. Rodean, *Stochastic Lagrangean Models of Turbulent Diffusion* (American Meteorological Society, Boston, 1996), 84 pp.

7. A. Stohl, Atmos. Environ. 32, No. 6, 447-466 (1998).

8. R.R. Draxler and G.D. Hess, *Description of Hysplit 4 modelling system*. NOAA Technical Memorandum ERL-224 (1988), 24 pp.

9. R.R. Draxler, and G.D. Hess, Austral. Meteorol. Magazine, No. 47, 295–308 (1998).

10. J.F. Pudykewicz, Atmos. Environ. **32**, No. 17, 3039–3050, (1998).

11. G.I. Marchuk, Mathematical Simulation in the Problem of Environment (Nauka, Moscow, 1982), 320 pp. 12. G.I. Marchuk, Conjugated Equations and Analysis of

Complicated Systems (Nauka, Moscow, 1992), 425 pp. 13. V.V. Penenko, Bull NCC: Num. Model in Atmosph.,

No. 4, 31–52 (1996).

14. V. Penenko, A. Baklanov, and E. Tsvetova, FGCS 18, 661–671 (2002).

15. V.V. Penenko and E.A. Tsvetova, Atmos. Oceanic Opt. **12**, No. 5, 447–449 (1999).

16. E. Kalnay, M. Kanamitsu, R. Kistler, et al., Bull. Am. Meteorol. Soc. 77, 437–471 (1996).

17. A.I. Borodulin, A.S. Safatov, V.V. Marchenko, A.N. Shabanov, B.D. Belan, and M.V. Panchenko, Atmos. Oceanic Opt. **16**, Nos. 5–6, 387–390 (2003).

18. A.I. Borodulin, G.A. Maistrenko, and B.M. Chaldin, Statistical Description of the Process of Turbulent Diffusion of Aerosols in the Atmosphere (State University Publishing House, Novosibirsk, 1992), 124 pp.