TEMPORAL AND SPATIAL VARIABILITY OF FIELDS OF THE OPTICAL AND AEROSOL CHARACTERISTICS IN THE ATMOSPHERE. II. AEROSOL CHARACTERISTICS

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The paper describes the results of application of the normalized span method to an analysis of the observed aerosol characteristics – the number density and differential number density of aerosol particles as well as their size distribution function. The values of the Hurst constant for the considered microstructural characteristics depend significantly on the time scale that enables us to discuss the nature of processes determining their variations. The number density of aerosols and their dispersed composition in the ground layer are shown to have regular daily behavior, as it also follows from the results of traditional microphysical measurements, but the regularities of their variability for some ranges of particle sizes differ significantly.

In Ref. 1 the results of application of the normalized span method to an analysis of several samples of the experimental data on the optical characteristics of the atmosphere were considered. The basic conclusion of Ref. 1 can be briefly formulated as follows: temporal variations of the atmospheric optical characteristics – the values of optical thickness or the extinction coefficients calculated on their basis – obey the Hurst law with the Hurst constant H that differs significantly from $H = 0.5$, which testifies to the difference of the processes, determining the time variability of the optical characteristics, from random ones. According to Ref. 1, the variations of the atmospheric optical characteristics completely coincide with the variations of their aerosol component (in Ref. 1, the data were considered for the visible and near-infrared ($\lambda = 2.2$ μm) spectral ranges). This can be treated as one more evidence of the determining role of aerosols in the formation of optical properties of the atmosphere in these spectral ranges or as an evidence of the fact that the aerosol state of the atmosphere is governed by the same laws as the atmospheric stratification, its molecular composition (above all, the water vapor content), and spatial distribution of radiation-absorbing gases.

The optical characteristics of aerosols, including time variations of the aerosol extinction analyzed in Ref. 1, are complex functions of many parameters describing the microstructure and the composition of particles. It is rather difficult to identify the parameters whose variations result in the variations of the observed optical characteristics. We also can assume that the spatial distribution of atmospheric aerosols has a complex microstructure: against the background field of relatively homogeneous aerosol particles of similar composition (background aerosols), the cloud-like formations of particles with different microstructure drift. In accordance with the present-day concepts on regularities of formation and evolution of such systems,[2] we expect that the structure of such a field obeys the laws of fractal geometry.

From numerous characteristics of aerosol microstructure the number density and differential number density of particles are most accessible to measurements (as a rule, in a limited range of particle size).

The Laboratory of Aerosol Physics has at its disposal the unique data arrays. The above-mentioned measurements have been carrying out in the region of Old Peterhof since the foundation of the University Campus in 1976. They are carried out every year, as a rule, in summer (May–July) and fall (September-October) using the AZ-5 M photoelectric particle counter, which records the number density of aerosol particles in several ranges of particle sizes for $d \geq 0.2$ μm. (The endpoints of these ranges are shown in Fig. 1 in which the examples of measurements are illustrated. Note here that the values of d given in the text correspond to the rated data of the device and are for the most part conventional. Accurate determination of the endpoints of the ranges of particle sizes is a very complicated metrological problem and requires special discussion.) The maximum size of recorded particles is about $d \approx 15$ μm (larger particles have a very low aspiration coefficient). The error in measuring the number density of particles with moderate sizes is $\delta N/N \approx 15\%$. It rises sharply in the range of particle sizes $d \geq 3.5$ μm because of very poor statistics (in this range of particle sizes the statistical measurement error is within the limits from 30 to 100%).

The measurements with the photoelectric counter, as a rule, consist in periodic sampling of aerosols on

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the Petryanov filters with subsequent laboratory analysis (of dispersion and elemental composition of samples). Unfortunately, the observation periods are short, as a rule, they are 15–20 days. This short duration of individual observation cycles does not permit the long-term aerosol variability to be analyzed in details.

In settlement Dubochki (on the southern shore of the Gulf of Finland 15 km to the west of the Old Peterhof University Campus, where a greater part of the data array was obtained) in the course of integrated aerosol and optical investigations[3] the number density of the near-ground aerosol was measured in June 1994. The number density and particle size distributions were measured every hour from 9 a.m. till 11 p.m. from June 3 till July 3, 1994, every day (of course, without heavy rain). The measurements were regularly made during 2 or 3 days. The data of this type were used to analyze the diurnal aerosol variability. The experiments used for analysis contain 40–50 series of measurements. (Unfortunately, towards the end of the above-mentioned period the thunderstorms began that did not enable us to carry out completely the projected observations.)

It should be noted that although the samples available are limited and this might cast some doubt on the reliability of the results of their statistical processing, such duration of individual cycles, taking into account the weather conditions in the Leningrad region during the observation period, ensures relative stability of air mass over the entire measurement cycle. In other words in this case the variations of the total number density and aerosol size distribution are conditioned primarily by local processes (the effect of aerosol sources and sinks located in the immediate vicinity of the measurement station) and by such processes as diurnal change of atmospheric stratification and convective flows.

A comparison of measurements[3] and the analogous data obtained for the entire period of measurements in the Old Peterhof region shows that general regularities of variation of particle number density and diurnal transformation of the particle size distribution function as well as the data on the aerosol particle composition are in good agreement with the corresponding characteristics averaged over a period of many years. These characteristics can be considered typical of the given region and season. At the same time, when analyzing the results,[3] the following peculiarities were found resulting in their deviation from the typical data of long standing:

1) The obtained particle size distributions were nonstandard and, evidently, deformed strongly by many coarse-dispersed particles presented in the air that were evidently of organic (vegetative) origin.

2) As a rule, we observed the sharp decrease of $N(d)$ values in the range $d \approx (0.4–0.6)$ μm, which is indicative of nonstationarity of particle size spectrum evolution (the occurrence of adjacent intermittent particle sources).

3) Flocculent (cloudy) spatial structure of aerosol field is often observed, which is clearly manifested in a series of successive measurements of particle size distribution and their number density for practically continuous observations in the fixed range of sizes. The recorded inhomogeneities of spatial aerosol distribution are relatively small by scale (several tens or hundreds of meters). However, frequently observed sharp deviations of microstructural characteristics from the mean ones for the time intervals up to 1–2 h are indicative of the occurrence of larger-scale inhomogeneities of the aerosol structure.

On this basis, to analyze the structure of time series of measurements of aerosol particle size distribution and particle number density, the following data arrays were formed:

I) Observations of diurnal behavior on June 7–9, 1994 (Dubochki, 47 series of observations, the time interval was Δt = 1 hour).

II) Observations of diurnal behavior on June 11–13, 1994 (Dubochki, 42 series of observations, the time interval was Δt = 1 hour).

III) Observations of diurnal behavior on June 6 – July 3, 1992 (Dubochki, 141 series of observations, the time interval was Δt = 1 hour). This data array included arrays I and II as well as the data obtained on June 20–21, June 30–July 1, and July 2–3 when the diurnal observations were interrupted due to showers or for technical reasons (disconnection from the power supply).

IV) Observations in June–July, 1994 (Dubochki, Old Peterhof, 67 series of measurements, the time interval was Δt = 12 hours). This data array comprised the measurements over the above-indicated period in Dubochki and near the building of the Scientific Research Institute of Physics at the St.-Petersburg State University in Old Peterhof. Taking into consideration significant diurnal variations of measurable parameters (see Fig. 1), only the observations between 9–10 and 18–20 hours were processed.

Tables I–III present the results of analysis by the method of normalized span of experimental data on the short-term variability of aerosols.

Summarizing the results of experimental data processing, the following should be noted:

1. When analyzing the data on the number density of particles whose sizes exceed d_0 or are within the definite range (see Tables I and II) we should focus our attention primarily on the different values of the Hurst constant H attendant to variations of the time interval: if at Δt = 1 hour (data arrays I, II, and III) the values of H, as a rule, are small ($H \le 0.3$), then at Δt = 12 hours (data array IV) $H \ge 0.5$. This evidently suggests that the particle number density on different time scales is governed by different processes. (Similar situation takes place for the observations of the aerosol component of atmospheric optical thickness.[1])

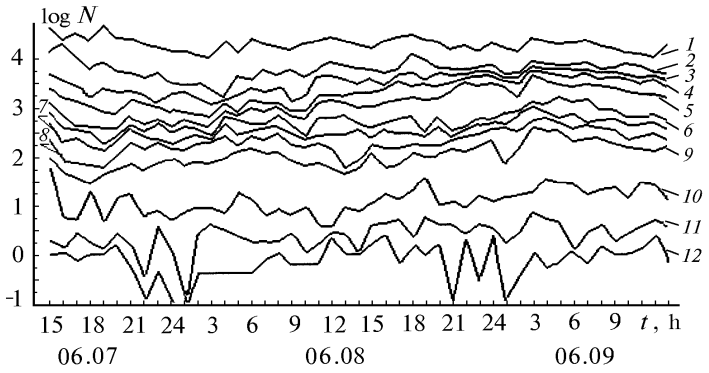

FIG. 1. Temporal variations of the number density of aerosol particles with sizes exceeding d_0 = 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.5, 2, 4, 7, and 10 μm *(curves 1–12, respectively).*

TABLE I. The Hurst constant H for the aerosol particle number density N ($d > d_0$, μm).

d_0, μm	I	II	III	IV
0.4	0.212±0.007	0.207±0.006	0.21±0.01	0.62±0.05
0.5	0.255±0.007	0.248±0.007	0.25±0.01	0.67±0.06
0.6	0.229±0.010	0.235±0.009	0.23±0.01	0.67±0.07
0.7	0.282±0.012	0.277±0.010	0.28±0.01	0.70±0.07
0.8	0.331±0.013	0.340±0.011	0.34±0.01	0.76±0.07
0.9	0.303±0.013	0.311±0.012	0.31±0.015	0.75±0.07
1.0	0.352±0.02	0.360±0.015	0.36±0.02	0.75±0.08
1.5	0.380±0.03	0.392±0.02	0.39±0.04	0.78±0.07
2	0.445±0.04	0.450±0.04	0.45±0.05	0.58±0.09
4	0.320±0.03	0.350±0.04	0.34±0.04	0.62±0.08
7	0.355±0.04	0.362±0.03	0.36±0.04	0.60±0.08
10	0.368±0.04	0.380±0.03	0.37±0.04	0.61±0.09

TABLE II. The Hurst constant H for the differential number density of aerosol particles $\Delta N/\Delta d$.

Δd, μm	I	II	III	IV
0.4–0.5	0.182±0.008	0.207±0.006	0.20±0.02	0.60±0.04
0.5–0.6	0.202±0.006	0.218±0.007	0.21±0.02	0.68±0.06
0.6–0.7	0.104±0.004	0.085±0.009	0.10±0.015	0.63±0.06
0.7–0.8	0.082±0.007	0.077±0.010	0.08±0.01	0.72±0.06
0.8–0.9	0.053±0.008	0.048±0.011	0.05±0.01	0.74±0.07
0.9–1.0	0.031±0.006	0.043±0.012	0.04±0.015	0.73±0.07
1.0–1.5	0.048±0.007	0.060±0.015	0.05±0.02	0.76±0.08
1.5–2	0.069±0.009	0.082±0.010	0.08±0.03	0.73±0.07
2–4	0.045±0.008	0.045±0.015	0.05±0.04	0.56±0.09
4–7	0.066±0.006	0.053±0.012	0.06±0.03	0.60±0.08
7–10	0.055±0.012	0.062±0.011	0.06±0.03	0.58±0.08
10–15	0.055±0.014	0.068±0.018	0.07±0.04	0.57±0.09

It should be noted that $H \to 0$ for periodic (or close to periodic) variations of the observed parameter. Thus, evidently the variations of particle number density undergo regular diurnal fluctuations. When the observation time interval is comparable with the variation period or exceeds it, such process cannot be detected.

2. When analyzing the data on the normalized function of particle size distribution (Table III) for all considered data arrays, the values of the Hurst constant turned out to be small (in many cases, their difference from zero is comparable with the calculation error). The physical sense of such a result is not quite clear. It is possible that this is connected with the fact that during observations in Dubochki (data arrays I, II, and III) the distribution function varied regularly during 24 hours (in Ref. 2, in particular, the diurnal variations of the median radius are analyzed), and when these data were combined with the results of observations in Old

Peterhof (data array IV), the point spread on the plot increased markedly (note that in the plots of the normalized span dependence on time used to determine the values of the Hurst constants for the data arrays III and IV abrupt changes were regularly observed).

3. In Table I the fact has engaged our attention that values of H for the channel $d \geq 2$ μm (data arrays I, II, and III) and for the channels $d \geq 2$, 4, 7, and 10 μm (data array IV) are close to $H = 0.5$. This may indicate either the anthropogenic nature of particles in the corresponding ranges of particle sizes or the large measurement error in the given channels (recall that the error of measuring the number density of giant particles may reach 30–100%). In this case, the measurement error may be so large that the results contain practically no information on the aerosol. However, a comparison of the data in Tables I–III makes the first assumption preferable.

TABLE III. The Hurst constant H for the normalized function of aerosol particle size distribution $\Delta N/(N \cdot \Delta d)$.

Δd, μm	I	II	III	IV
0.4–0.5	0.112±0.007	0.097±0.006	0.11±0.01	0.22±0.02
0.5–0.6	0.105±0.007	0.086±0.007	0.09±0.01	0.17±0.02
0.6–0.7	0.079±0.010	0.065±0.009	0.07±0.01	0.12±0.07
0.7–0.8	0.082±0.012	0.077±0.010	0.08±0.02	0.11±0.07
0.8–0.9	0.031±0.013	0.054±0.011	0.05±0.02	0.11±0.07
0.9–1.0	0.033±0.013	0.053±0.012	0.05±0.015	0.07±0.05
1.0–1.5	0.042±0.02	0.060±0.015	0.06±0.02	0.07±0.06
1.5–2	0.038±0.017	0.039±0.02	0.04±0.03	0.08±0.07
2–4	0.045±0.025	0.045±0.04	0.05±0.05	0.08±0.08
4–7	0.025±0.020	0.053±0.04	0.05±0.04	0.06±0.05
7–10	0.075±0.035	0.062±0.030	0.06±0.04	0.07±0.05
10–15	0.097±0.040	0.080±0.035	0.09±0.05	0.10±0.09

4. In Ref. 1 it is shown that the curves of the dependence of the normalized span on time for atmospheric attenuation and its aerosol component practically coincide, while the analysis of corresponding regularities of the microstructural characteristics yields significantly different estimates of the Hurst constants. However, we believe that there are no contradictions in these estimates, because, on the one hand, cyclicity of variations of the optical and microstructural aerosol characteristics manifests itself identically in Ref. 1 (days with standard conditions) and in this case. On the other hand, the variations of the aerosol characteristics in the ground atmospheric layer considered here undoubtedly are very important for the formation of optical characteristics of the atmospheric column but hardly determine it completely. In this case, the characteristics of the underlying surface and local ground-based aerosol sources are most clearly manifested, while for observations of the atmospheric column their contribution is much less.

REFERENCES

1. S.D. Andreev and L.S. @vlev, Atm. Oceanic Opt. **10**, No. 12, 900–905 (1997).
2. F. Feder, *Fractals* [Russian translation] (Mir, Moscow, 1991), 260 pp.
3. S.D. Andreev, V.M. Zhukov, L.S. @vlev, et al., in: *Abstracts of Reports at the Second Interrepublic Symposium on Atmospheric and Oceanic Optics*, Tomsk (1995), pp. 95–96.