ON THE TRANSFORMATION OF THE PARTICLE SIZE-SPECTRUM OF AN AEROSOL DUE TO A CHANGE IN THE HUMIDITY

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The changes in the number density of aerosol particles are studied based on data from airborne measurements of the particle-size spectra of the aerosol for different values of the relative humidity. It is shown that the variation of the number density exhibits regional features. This is determined by the chemical composition of the aerosol particles.

It is well known that atmospheric aerosols play an important (if not the main) role in the variation of the optical properties of air and thereby determine the Earth's radiation budget. Being distinguished by exceptional diversity of their properties, the aerosol particles, in their turn, are affected by variations in the physical characteristics of air. One factor determining the change in the microphysical and optical properties of aerosol is the change in the humidity. It is customarily assumed that in the real atmosphere an increase in the relative humidity gives rise to an increase in the particle size.[2] This dependence is usually written with the help of the formulas of F. Kasten or J. Hanel:[3,4] $a_f = a_c(1 - f_0)\varepsilon$, where $\varepsilon = 0.18–0.255$, depending on the type of aerosol; a_d is the radius of a dry particle; and, f_0 is the relative humidity.

The formulas in Refs. 3 and 4 have been confirmed many times based on data obtained in the layer near the ground, i.e. , near the underlying surface — the main source of aerosol, water vapor, and aerosol-forming vapors. Our airborne experiments on artificial wetting of aerosol have shown that in more than 90% of the realizations the dependence of the scattering coefficients on the relative humidity can also be approximated at all altitudes by a similar formula.[5]

As a result of investigations performed using the airborne laboratory of the Institute of Atmospheric Optics of the Siberian Branch of the Academy of Sciences of the USSR, a large volume of data on the particle-size distribution of aerosol with a wide range of values of the relative humidity has now been accumulated. The measurements were performed with the help of the instrumentation described in Refs. 6 and 8. A check of the formulas of Refs. 3 and 4 on our data showed that they did not always hold for the boundary layer and free atmosphere. For this reason the purpose of this work is to analyze the possible reasons for the breakdown of these formulas.

The microphysical properties of the aerosol were measured using an improved AZ-5 counter; the same counter was used for 7 years. Since the operation of the AZ-5 counter generates many remarks we shall first examine the methodical questions regarding the measurement of the characteristics of aerosol by the photoelectric method implemented in this counter.

The possible errors in the measurement of the particle-size distribution function of aerosol by photoelectric counters in a real atmosphere under conditions of variable humidity were examined in Refs. 9–11. The main factors affecting the measurement accuracy, according to the data of Refs. 9–11, are the deviation of the optical constants of the aerosol under study, primarily the complex refractive index of the particle material, from the corresponding characteristics of the aerosol employed for calibration (usually $m = 1.60–0i$), and the fact that there is a lower limit on the particle sizes that can be recorded by the counters. In the measurements the humidity can give rise to strong deviations from the calibration conditions (a change in the visibility S_m from 50 to 5 km and gives rise to a change in the real part of the refractive index from 1.50 to 1.36). The existence of strong absorption ($\kappa = 0.1$) can cause the real particle sizes to be underestimated by up to 600% (Ref. 10).

According to Refs. 10 and 11 the true particle sizes a^* is related with the standard size a (1, 6) by the relation

$$a^\bullet(n) = a(1.6)(0.6/n-1)^{1.2-0.261n\rho} \cdot 1.6, \quad (\rho = 2\pi a/\lambda),$$

where a is the particle radius and λ is the effective wavelength, equal to 0.6 μm. Specifically, for the AZ-5

$$a^\bullet(0.2; n) = a(0.2); 1.6)0.6/n-1,$$

$$a(0.2; 1.6) = 0.2 \text{ μm.}$$

Since the particle radius depends on the humidity $a(f) = a_0(1 - f)^{-\nu}$, where a_0 is the radius of a dry particle; f is the relative humidity in hundredths; and, γ is a condensation activity parameter, when the substance dissolves the refractive index of the particle

material $n(f)$ also changes:

$$\frac{n(f) - n_w}{n_0 - n_w} = (1 - f)^{3\gamma},$$

where n_w is the refractive index of water.

According to Ref. 9 the counter can respond in two ways.

1) As the particle size increases with the humidity increase the distribution function will shift to the right along the size scale and the AZ-5 counter will record an increasingly larger number of particles. Therefore, for a constant lower limit of measurable particle size (a_0), the counter must record the increase in the measuredparticle number density N^* as f increases (if the total particle number density is constant).

2) As the humidity increases, $n(f)$ decreases and, as a consequence, the lower limit of particle sizes (a_0) recorded by the counter will shift to the right along the size scale, and this should result in a decrease in N^* with the total particle number density remaining constant.

It follows from the physics of aerosol that the process of wetting the aerosol and the dissolution of the aerosol material do not occur instantaneously. For this reason three results of the interaction of the two mechanisms described above are possible:

1) If the rate of displacement of a_0 is less than the rate of displacement of dN^*/da, the counter gives an increase in number density;

2) If the rate of change of a_0 is equal to that of dN^*/da, $N^*(f)$ is constant;

3) If the rate of displacement of a_0 is greater than that of dN^*/da, the counter records a decrease in N^*.

The calculations performed in Refs. 10 and 11 showed that for all values of the condensation activity γ the counter should record a monotonic increase in the total particle number density $N (r \geq a_0)$ with increasing humidity.

In Refs. 9 and 10 the following formula was proposed to correct measurements:

$$N = N^*[0.5 \pm \Phi(t)], \quad (a_0 \geq a),$$

where a is the recorded aerosol particle; $\Phi(t)$ is the error integral and $t = \ln\left(\frac{a_0}{a}\right)\Big/ S$, and the parameter S takes into account which the polydispersed nature of the aerosol and the fact that the mode of the distribution is dispersed from the fixed position of the lower limit of the counter a_0.

The dependence $N^*(t)$ is approximated with high accuracy ($\pm 2\%$) by a parabola in a wide range of values of the parameter $t = -2.8 + 0.5$

$$\ln(N^*/N) = \alpha t^2 + bt + c,$$

where $a = -0.394$, $b = 0.773$, and $c = -0.675$.

To determine the trend of the changes in the indications of the counter, as the humidity, increases the entire block of data from measurements of the aerosol particle distribution over the spectrum (more than 3000 spectra for Western Siberia and more than 1000 spectra for Kazakhstan) was divided into four groups: $f \leq 30\%$ (I), $f = 31–59\%$(II), $f = 60–79\%$ (III), and $f \geq 80\%$ (IV). For each group the average values were calculated irrespective of the altitude and conditions andthe spectra of the aerosol particle-size distribution, which are shown in Fig. 1.
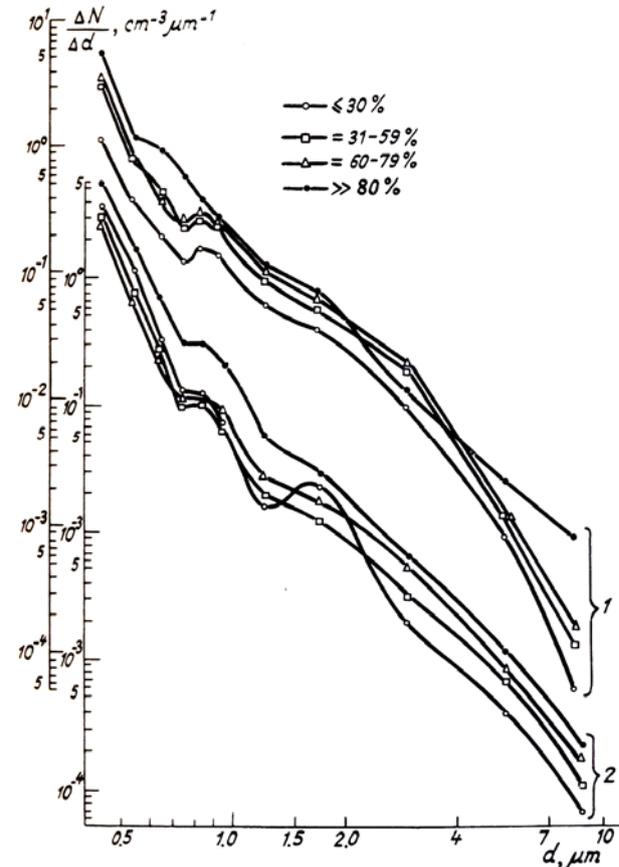
FIG. 1. The aerosol particle distribution according to the spectrum above Kazakhstan (1) and Vestern Siberia (2) with different humidity.

One can see from Fig. 1 that for the region of Kazakhstan as the humidity increases (the group of curves 1) there is a tendency for the aerosol particle concentration to increase over the entire section of the particle-size spectrum studied. Therefore wetting of the particles and an increase in their size results in more rapid displacement of the entire distribution function as a whole toward the right, while displacement of the lower limit a_0 owing to the decrease in the refractive index n occurs more slowly. As the humidity changes the concentration of the submicron fraction increases by a factor of 5 and almost by a factor of 10 — coarsely dispersed fraction. The vertical distribution of the aerosol number density for three fractions is shown in Fig. 2, whence one can see that an increase in the humidity results in an increase in the number density in almost the entire range of altitudes studied.
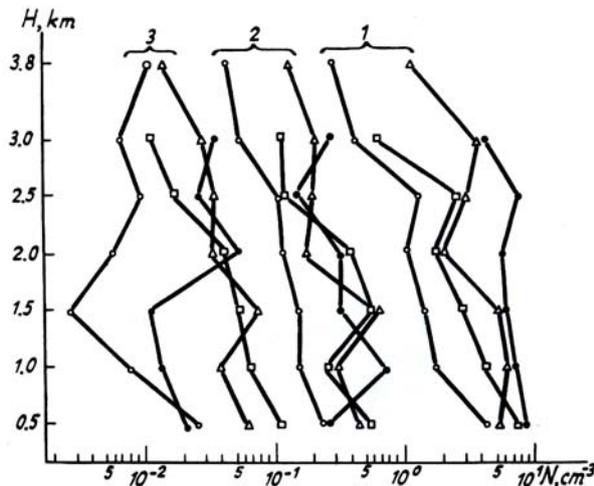
FIG. 2. The vertical distribution of the aerosol above Kazakhstan for the fractions $d = 0.4–0.5$ µm (1), $d = 0.8–0.9$ µm (2), and $d = 4–7$ µm (3) with different humidity (see FIG. 1 for an explanation of the labeling).
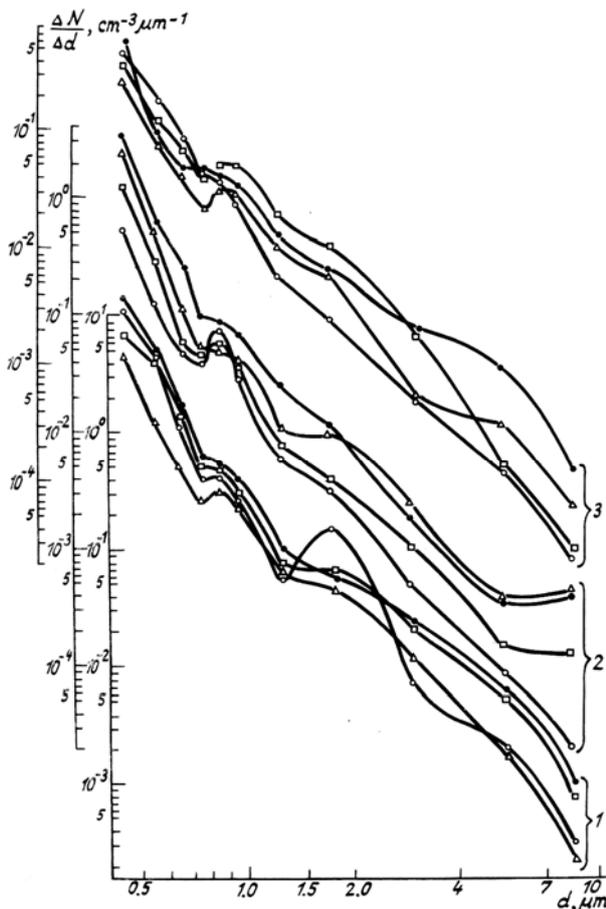
FIG. 3. The distribution of aerosol particles ever the spectrum above Western Siberia at altitudes of 600 m (1), 2500 m (2), and 4000 m (3) for different values of the humidity (the labeling is explained in Fig. 1).

For Western Siberia this picture is somewhat more complicated (Fig. 1, the group of curves 2). If the coarsely dispersed fraction exhibits the same behavior, i.e., an increase in the humidity results in an increase in the measured number density, then the behavior of the submicron fraction depends on the value of the relative humidity: as the relative humidity increases up to $f = 60–79\%$ the number density for the submicron fraction at first decreases and then with humidity $f \geq 80\%$ it increases rapidly. The analysis performed for the aerosol particle spectra measured at different altitudes with different values of the humidity shows (Fig. 3) that the characteristic behavior observed does not occur for all of them. If the above-described feature is present at an altitude of 600 m (the group of curves 1), then at an altitude of 2500 m the change in the number density with increasing humidity follows a dependence of the Kasten-Hanel type (the group of curves 2); at an altitude of 4000 m (the group of curves 3) the trend toward increasing number density as a function of the humidity once again breaks down for the submicron fraction.

Such differences in the measurements of the aerosol number density for different values of the relative humidity above two regions can, in our opinion, be explained by the difference in the chemical composition of the aerosol; this was also pointed out in Ref. 11. As a confirmation of this idea we shall present data on the chemical composition of the aerosol above both regions (Table I).

It follows from the data given in Table I that above Western Siberia ions and elements characteristic for particles of photochemical origin, which are usually less than 1 µm in size, i.e., they fall into the submicron fraction, predominate in the composition of the aerosol. These are the ions SO_4^{-2}, Cl^-, NO_3^-, and K^+. Conversely, above Kazakhstan elements which are characteristic for coarsely dispersed particles originating from the soil Al, Ca, Si, Fe, and Mg are present in the aerosol. Since the chemical composition of the aerosol determines the microstructure of the particles and therefore the mechanism of wetting and dissolution of the material this is reflected in the dynamics of the number density of the submicron fraction.

In our opinion the dynamics of wetting of aerosol above Western Siberia can be explained as follows. At the starting stage of wetting of the aerosol particles the particle material is dissolved, which results in a decrease in the refractive index n and correspondingly to a displacement of a_0. When the relative humidity exceeds 80% condensational growth of the particles starts to predominate, dN^*/da increases rapidly, and correspondingly the measured aerosol number density N^* increases.

TABLE I.

The chemical composition of aerosol above Western Siberia and Kazakhstan ($\mu g/m$): numerator — average value; denominator — number of samples.

Region	Element					Element					
	Si	Fe	Mg	Na^+	K^+	SO_4^{2-}	NH_4^+	NO_3^-	Cl^-	Al	Ca
Western Siberia	3.57/221	1.30/225	0.18/224	0.34/236	0.09/235	0.44/234	0.22/233	5.95/234	3.01/235	2.04/286	0.42/146
Kazakhstan	17.64/126	2.81/135	0.73/86	0.92/205	0.02/203	0.28/203	0.32/204	3.01/203	1.15/201	3.29/138	0.67/69

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