INVESTIGATION OF THERMO– AND HYGROOPTICAL CHARACTERISTICS OF ATMOSPHERIC AEROSOL ABOVE THE ATLANTIC OCEAN

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The results of investigation of aerosol in the atmospheric layer adjacent to water above the Atlantic Ocean are presented. The measurements were made using a photocounter of particles and a nephelometer equipped with thermo– and hygroactuators of aerosol. The influence of the continent on formation of optical and microphysical properties of aerosol above the ocean is analyzed. A variation in proportion of submicron and medium–sized particles is shown to be followed with a law by a change of thermooptical parameters of aerosol. A good reproducibility of these parameters for marine aerosol is noted.

Following the long–term program of investigating the atmospheric aerosol in different geographic regions the measurements were carried out in the central and northern parts of the Atlantic Ocean in April and June, 1988 during the 9th mission of the research vessel "Prof. Sergei Dorofeev" of Leningrad Hydrometeorological Institute. The course of the vessel is depicted in Fig. 1.

The goal of the experiment was to obtain information about thermo– and hygrooptical characteristics of aerosol[1] in the atmospheric layer adjacent to water and to estimate the effect of the continent on formation of composition and properties of aerosol particles.

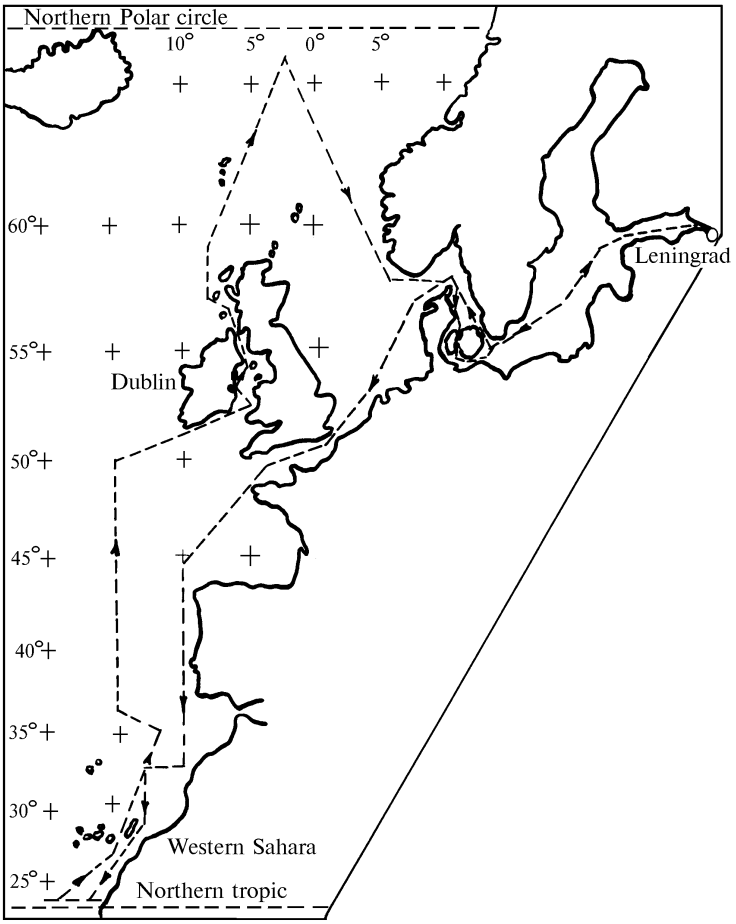

Fig. 1. Diagram of the 9th mission of the vessel "Prof. Sergei Dorofeev".

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Based on the available notion of the effect of land on dispersed composition of air over the ocean,[2–6] it is possible to assume that its action must be manifested in two ranges of particle size: a submicron region, where particles of photochemical origin are concentrated, including anthropogenic ones, and a range of medium–sized fraction with terrigenous particles. Above considerations determined the choice of optimal local meters: a nephelometer for submicron particles and an AZ–5 photocounter that operates best in the mean–dispersed size range.

The nephelometer, constructed on a base of commercial FAN photometer and equipped with a moistener and heater for the aerosol, has been detailed in Ref. 1. Moistening of aerosol with the control over relative humidity of air in an operating cell of the nephelometer enable us to calculate the index of condensation activity γ for the coefficient of directed scattering $\mu(45°)$ using the Hanel empirical formula.[7] The temperature parameters F and Q determined as a ratio of the coefficients $\mu(45°)$ at a fixed temperatures are found from the temperature dependence of $\mu(45°)$.

The setup was mounted in a wheel–house of the vessel. The aerosol was taken in an incoming air flow with forced transportation to an operating volume through a plastic tube 8 m in length and 20 mm in an inner diameter. The air consumption in the operating volume of the setup was 20 to 30 liters per minute. An inlet of the air intake was at an altitude of about 8 m above the water surface on a windward board. To do this the air intake could be taken from one board to the other depending on the vessel orientation. The nephelometer measurements were basically made by day with a 4 hr interval. An interval between microstructural measurements with an AZ–5 was from 2 to 8 hrs. About 200 measurements were carried out during the mission. The mean values and their rms errors for nephelometric data obtained during the mission are listed in Table I.

TABLE I.

	Parameter						
	$\mu(45°)$, $km^{-1}\ sr^{-1}$	$P(90°)$	γ	F	Q	RH, %	W, m/s
Mean value	0.011	0.13	0.30	1.34	0.47	80.5	6.5
Rms error	0.011	0.11	0.11	0.17	0.20	8.3	3.2

Here $\mu(45°)$ is the coefficient of directed scattering at an angle of 45°; $P(90°)$ is the degree of polarization of scattered radiation at an angle of 90°; RH and W are the relative humidity of air and wind velocity, respectively; and, F and Q are the thermo–optical parameters determined from the following relations: $Q=\mu(250°C)/\mu(100°C)$; $F=\mu(T+15°C)/\mu(100°C)$, where T is the temperature of ambient air.

It is believed that at temperature 15° higher than that of the ambient air the aerosol occurs practically at zero relative humidity of air; at 100°C the whole of bound water is removed from composition of particles; while at 250°C only NaCl and soot remain undecomposed.

It should be noted that our data were obtained under high transmittance of the atmosphere and meteorological parameters specific for a smooth ocean. At the same time, averaging of data separated in space and time makes the processes of formation of an optical situation in the atmosphere obscure. Therefore, let us analyze some portions of the data set separately.

Figure 2 depicts spatiotemporal sections of directed scattering coefficients observed when moving away from the continent or approaching it. It was observed that moving away from industrial centers of Europe results in reduction of the scattering coefficients by a factor of more than five. It could be accounted for by variations in meteorological and synoptic situations (a period of more than 10 days). However, this pattern was constantly reproduced in shorter cycles of measurements as well (Fig. 2*b* and *c*).

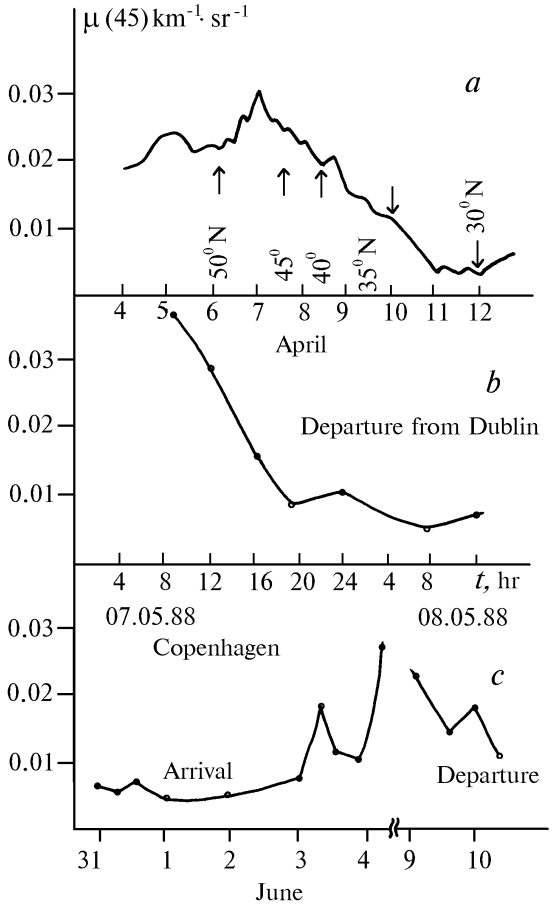

Fig. 2. Spatiotemporal sections of the directed scattering coefficients $\mu(45°)$.

The analysis of microstructural data shows that the scattering coefficient, when moving away from the continent, decreases due to decrease of absolute and relative content of submicron particles. As seen in Fig. 3, where the averaged normalized functions of particles size distribution over volumes are represented for different regions of Atlantic, the contribution of continental air is pronounced in the submicron range of the spectra obtained near the ocean coast, while the aerosol particles size spectra in the open ocean are typical for most of cases.

Particles of a medium–sized fraction which were observed in the open ocean came to the atmosphere from the sea surface as a result of the well–known bubble mechanism of particle formation, and their content is primarily caused by the extent of the sea surface roughness and wind velocity.[2–4] Our experimental data agree with this concept. To illustrate the effect of the wind velocity W on the increase of medium–sized particle concentration, Fig. 4 depicts particles volume distributions observed at a stable weather during a day.

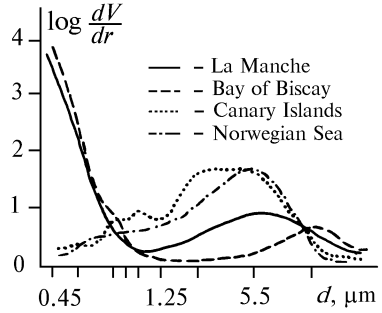

Fig. 3. Averaged normalized functions of particles distribution over volumes for different regions of Atlantic: La Manche (solid line), Bay of Biscay (dashed line), Norwegian Sea (dot-and-dash line), and Canary Islands (dotted line).

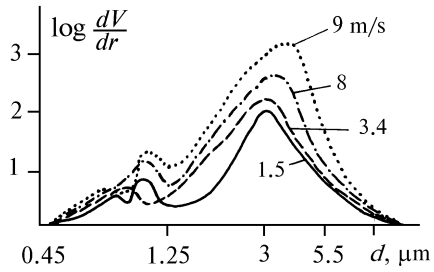

Fig. 4. Volume distributions of particles for different wind velocities.

Among the analyzed thermo– and hygrooptical characteristics, the increase of relative content of marine aerosol (and, correspondingly, substantial decrease of submicron particles) was the most pronounced for the Q parameter. Since Q characterizes a portion of nonburning up (nondecomposable) residue at ~ 250°C, we can assume that the increase of relative content of particles of salt origin must result in the increase of this parameter.[8] Actually, in our experiment such an increase was observed and it is depicted in Fig. 5*a* and *b*, where Fig. 5*a* is related to the section of μ(45°) in Fig. 2*a*, and Fig. 5*b* is related to Fig. 2*b*. A portion of nonburning up residue increases from 10–20 to 40–80%. Therefore, there is a reason to think that in oceanic observations the parameter Q can be used for estimating a portion of continental (in our case submicron) and marine particles.

Our concepts about volatility of submicron particles are in good agreement with the results of the authors of Ref. 8, who examined the temperature transformation of the particles size distribution function under maritime conditions. At the same time it was found that during the measurements near African continent some contribution to the increase of the parameter Q was also made by medium–sized particles of dust origin which were brought to the atmosphere over the ocean from Sahara Desert.

To show that the most volatile substances are characteristic of submicron particles (most probably, of nonmarine origin), we consider the situation observed on Canary Islands in more detail. Under the conditions of almost complete calm and very high transmittance of the atmosphere ($\sigma \sim 0.08$ km^{-1}) with the air flow from Canary Islands in the region of measurements we observed the increased concentration of finely dispersed particles (this situation was similar, to some extent, to the moments of formation of photochemical hazes which were observed before under continental conditions). The temporal section of this situation is shown in Fig. 6. For convenient comparison the data on μ(45°), particle concentration, and hydro– and thermooptical characteristics are given in relative units. As seen in this figure, the parameter F increases and the value Q decreases with the increase of the content of finely dispersed particles (increase of μ(45°)).

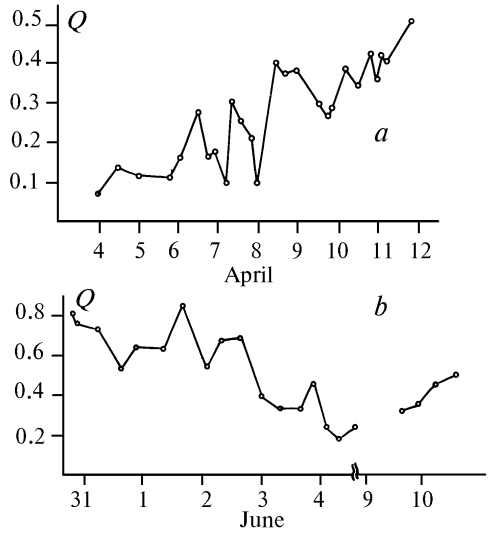

Fig. 5. The dependence of values of the Q parameter on changes of relative content of salt particles when moving away from the continent.

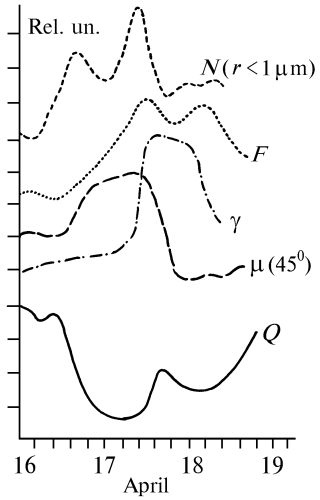

Fig. 6. Illustration of formation and transformation of photochemical haze over the ocean in the region of Canary Islands. $Q_{min} \sim 0.14$, $F_{min} \sim 1.20$, $\gamma_{min} \sim 0.25$, $Q_{max} \sim 0.36$, $F_{max} \sim 1.84$, and $\gamma_{max} \sim 0.60$.

The behavior of concentration of particles of $r < 1$ μm indicates that this process is related to finely dispersed particles (concentration of particles of $r > 1$ μm during this period was low and did not change). It is interesting to note that as this aerosol formation develops with maximum F we observe also maximum values of the parameter of condensation activity γ ($\gamma_{max} \sim 0.6$), while during oceanic measurements this was observed rarely. After F and γ become maximum (see Fig. 6) they keep

these values during several hours even though the total content of submicron particles decreases that is revealed from the decrease of $\mu(45°)$. This can be related to spatial spread of the formed haze.

At first glance small values of γ regularly recorded over the ocean are somewhat unexpected. However, this can be explained by the following considerations: the Hanel empirical formula[7] relates increasing particles size to the increase of relative air humidity RH, i.e.,

$$r = r_0 (1 - RH)^{-\varepsilon},$$

where r is the particle radius and ε is the parameter of condensation activity for r. In the range of submicron particles of radius $r \sim \lambda$ the scattering coefficient σ is proportional to r^3 (Ref. 5), and for $r \gg \lambda$ $\sigma \sim r^2$. Consequently, $\gamma \sim 3\varepsilon$ for submicron and $\gamma \sim 2\varepsilon$ for coarse particles. Thus, when the main contribution to the scattering coefficient formation is made by marine medium–sized particles it is possible to expect that the parameter of condensation activity γ for the scattering coefficient is related to the analogous parameter for a particle radius as $\gamma \sim 2.5\,\varepsilon$. The value $\varepsilon \sim 0.19$ was reported for sea salt particles.[7] But in Hanel's experiments the growth of sea salt crystals was investigated, while in a real atmosphere over the ocean there are particles originated from sea water droplets that cannot crystallize under specific relative humidity of sea air. In other words, these particles are in an upper part of the hysteresis loop of the RH dependence of r (Ref. 9). In this case the value of ε is smaller than 0.19. Moreover, we do not deal here with the volume coefficient of scattering σ but with the directed scattering coefficient $\mu(45°)$ where the normalized scattering phase function $f(45°)$ decreases with increase of relative humidity of air. Hence, the recorded values of the parameter of condensation activity $\gamma \sim 0.3$ are well explicable.

The continental air penetrating into the measurement regions in the open ocean on rare occasion, as a rule, resulted in the increase of the γ parameter. At the same time, a small number of observations of such situations which were usually accompanied with wind strengthening and, correspondingly, simultaneous increase of sea particle content, which conceal the exhibition of submicron particles, do not provide to justify this assumption and require further experimental tests be made using instrumentation for measuring particle dispersity in a submicron range.

As of the present time the effect of the continent on the content of coarse and medium–sized particles in the atmosphere over the ocean during dust removals from arid regions has been studied sufficiently well.[3] Under extremal conditions the dust removal changes strongly the optical situation in the atmosphere, and its contribution in the regions affected by this removal is predominant. Under ordinary conditions in these geographic regions it is difficult to separate out the contribution of dust particles to formation of an adjacent to water aerosol microstructure.

Our measurements in the regions of 24–25° north and 16–18° west recorded the increased value of the parameter $Q \sim 0.8$ that cannot be accounted for by a large number of sea salt particles under calm weather. To check the hypothesis on a contribution of dust particles precipitated from the upper level during the air flow directed from Africa, we made a spatial section along 24°N to the coast of Western Sahara. It should be noted that no substantial variations in the nephelometric data were observed, but a number of particles of $r > 1.5$ μm increased by a factor of more than four which is illustrated by a plot in Fig. 7 (here the distance from the coast is in Miles).

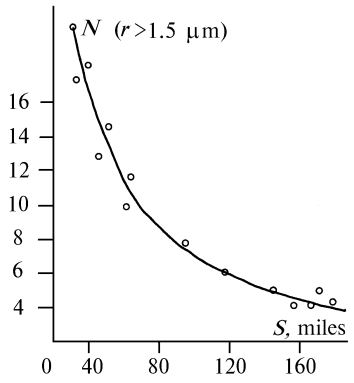

Fig. 7. Variations in dust particles content in the layer adjacent to water as a function of the distance from the coast of Western Sahara.

Focusing attention on the role of the continent in aerosol formation over the ocean, it also should be noted that the basic array of experimental data was obtained for conditions when the most of aerosol particles are certain to be of maritime origin. In these situations the thermo– and hygrooptical characteristics can be reproduced sufficiently well and their values are close to those listed in Table I.

To illustrate the agreement between the measurement results of thermo– and hygrooptical parameters in maritime hazes, Table II represents the data obtained in the contrast geographic regions of Atlantic which are outermost from each other (here the mean values of the parameters obtained under continental conditions are given for comparison too).

TABLE II.

Region of measurements	Parameter		
	γ	F	Q
Northern tropic	0.30	1.41	0.47
Northern Polar circle	0.32	1.31	0.49
Western Siberia	0.37	1.66	0.31

In conclusion, it can be pointed out that the effect of continental submicron particles is observed at sufficient distances from the coast and manifest itself both in absolute values of the scattering coefficients and thermo– and hygrooptical characteristics. The continental medium–sized particles in tropics of Atlantic can be of terrigenous nature and related to dust removal from Sahara Desert. In the open ocean, without the continent action, microstructural, thermo– and hygrooptical parameters are well reproduced and have certain differences from those observed on the continent.

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