Microphysical features of the aerosol component in different regions of the Atlantic

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We present some results of inverting the spectral data on the aerosol optical depth (AOD) measured over different regions of the Atlantic. It is shown that the remoteness of these regions from numerous secondary continental sources generating the aerosol is an important factor determining the features of the optical and microphysical properties of the atmospheric aerosol over the ocean. It has been found that in the mid-latitudes the volume content of the accumulative aerosol fraction $V_{ac}(r < 0.4\ \mu m)$ decreases by several times (from $3 \cdot 10^3$ to $3 \cdot 10^2\ \mu m^3 \cdot cm^{-2}$) with the distance from the European continent, while that of the medium fraction V_{md} ($0.4 < r < 1.2\ \mu m$) is stable and equal, on the average, to $1.4 \cdot 10^3\ \mu m^3 \cdot cm^{-2}$. In the tropics, the effect of intake of the continental aerosol manifests itself all over the size range analyzed. Just this effect governs the formation of the aerosol size spectrum. The results of interpretation of the mean spectral AOD have shown that V_{md} halves (from $12 \cdot 10^3$ to $6.0 \cdot 10^3\ \mu m^3 \cdot cm^{-2}$) when going on from the Sea of Darkness into the trade-wind zone, which confirms that the effect of the dust aerosol from the Sahara Desert weakens with the distance from the African continent. Despite the predominantly zonal distribution of the climate and meteorological characteristics of the atmosphere, the major factors determining the regional features of AOD over the ocean are the processes regulating the penetration of the continental air masses enriched with aerosol into one or another region.

Introduction

The study of the spectral transmittance of the atmosphere during five Atlantic research missions in 1989–1996 has yielded quite a representative data array on the variations of the aerosol optical depth (AOD).[1] It was noted that important roles in the formation of the spatial distribution of aerosol over the ocean belong to continental sources and the predominant type of air mass circulation in one or another region, which determine the direction and depth of the inflow of the continental aerosol into the maritime atmosphere.

To separate out the spatial component from the variations of the aerosol turbidity of the atmosphere, the Atlantic was divided into zones using two different approaches.[1,2] As a result, zones with significantly different AOD were determined. In the mid-latitudes the zones near continents (NC) (within several hundreds kilometers) and the zones of open ocean (OO) were isolated, while the tropical latitudes (partly the subtropics) were divided into the trade-wind (TW) zone, the zone of Sea of Darkness (SD), and the Canary Islands (CI) zone. The region near the equator on the periphery of the Gulf of Guinea was determined as an equatorial zone (EZ). The map of the zones is shown in Fig. 1.

For a more complete description of the aerosol properties in the entire column of the maritime atmosphere, this paper considers the features of the microstructure retrieved from the data of spectral measurements of AOD in different zones of the Atlantic.

Until recently, only few investigations of the maritime aerosol microstructure (based on the AOD inversion) have been carried out and, usually, at a short distance from the continents or in the coastal zone.[3–6] According to our classification, they fall in the same class, namely, the NC class. More precisely, it should be noted that our results obtained for the narrow coastal zone were not considered separately because of the paucity of the data acquired. With the development of the AERONET global network, regular investigations of the microphysical characteristics of aerosol have begun, in particular, at the island stations in different regions of the Global Ocean (in the Atlantic Ocean: Ascension Island, Bermudas, Canaries[6–10]). At the same time, the measurement results obtained on the islands correspond to a limited number of regions and can be overburdened by the impact of continental aerosol sources. Therefore, the investigations of aerosol using the data of shipborne AOD observations are still urgent as being more close to reality.

Technique of investigation

The disperse structure of aerosol was retrieved from the data on atmospheric AOD obtained with a multiwave sun photometer in the spectral region from 0.37 to 1.06 μm. The number of spectral intervals (wavelengths λ_i) in the course of the experiments increased from 4 in 1989 to 11 in 1995, 1996. For a more reliable retrieval of the microstructure characteristics, we used the most detailed data of the two latest missions. The error in AOD determination was estimated to be up to 0.02. In a more detail the characteristics of the photometer, the techniques of calibration and AOD determination have been considered in Ref. 11.

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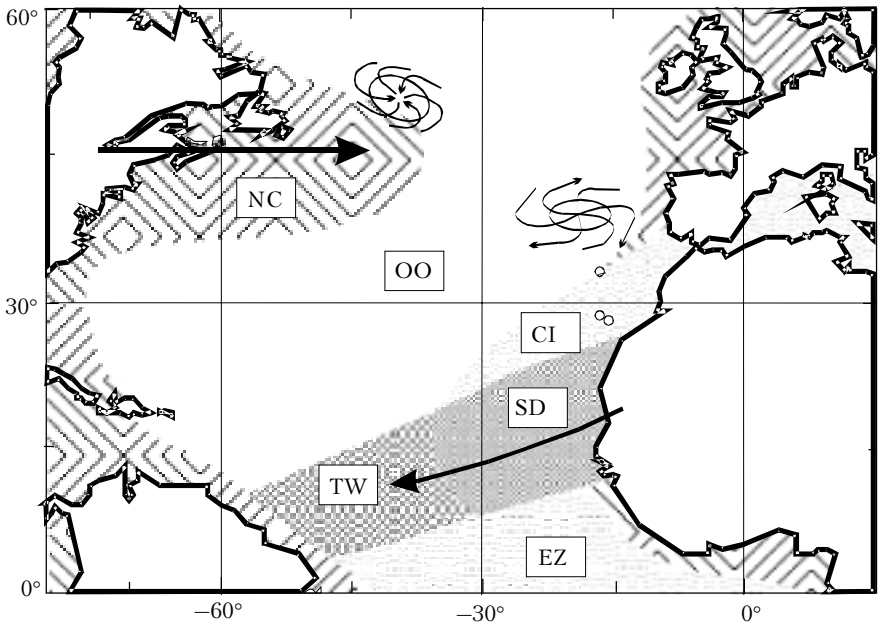

Fig. 1. Map of typical Atlantic regions according to the classification from Ref. 1.

The inverse problem was solved using the approach based on the Tikhonov's variational principle.[12] The features of the inversion technique are considered in detail in Ref. 13. The sought function of the particle size (cross section) distribution density $s_c(r)$ was determined from the solution of the system of equations of the following form:

$$\int_{R_1}^{R_2} K_\varepsilon(r,\lambda_i) s_c(r) \mathrm{d}r = \tau(\lambda_i),\ i = 1, 2, \ldots, n, \quad (1)$$

where $s_c(r) = \pi r^2 n_c(r)$; $n_c(r)$ is the particle size distribution in the atmospheric column; $K_\varepsilon(r,\lambda_i)$ is the efficiency factor of the aerosol extinction of radiation depending on the complex refractive index of particles $m = n - i\chi$; R_1 and R_2 are the boundaries of the distribution sought $s_c(r)$.

In inverting it was assumed that the real part of the refractive index is $n = 1.45$ and the imaginary part is $\chi = 0.005$. The upper estimate of R_2 was determined according to the technique proposed in Ref. 13 by the equation

$$(R_2)^{-1} \int_{R_1}^{R_2} K_\varepsilon(\lambda_{\max}, r)\, \mathrm{d}r = \bar{K}_m \tau(\lambda_{\max}) / \tau_{\max}, \quad (2)$$

where $\bar{K}_m = \tau_{\max} / S$ is the polydisperse extinction efficiency factor at the maximum of the spectral dependence $\tau(\lambda)$ and $S = \int_{R_1}^{R_2} s(r)\, \mathrm{d}r$. This technique requires the value of $\bar{K}_m$ to be specified *a priori* in accordance with the recommendations[13] from the range of 2.6–3.3. Equation (2) gives the lower estimate of R_2, that is, the true value lies to the right from the value found. In this connection, the estimate obtained from Eq. (2) was corrected toward the increase so that the total contribution of particles with the radii greater than R_2 to $\tau(\lambda)$ is negligibly small.

For example, for the regions with the increased turbidity of the atmosphere the estimates of the microstructure characteristics were obtained in the size range from 0.05 to 3.0 μm. In other words, the measurement data on AOD in the region $\lambda \sim 0.37$ to 1.06 μm bear information on the size distribution of particles with radii up to 2 to 3 μm depending on the particular realizations of the spectral profile $\tau(\lambda)$.

In the results obtained, the mode of the fine particles ($r < 0.4$ μm) within the size range of the accumulative fraction and one or two modes of large particles, which were denoted as the medium ($0.4 < r < 1.2$ μm) and coarse ($r > 1.2$ μm) modes, are quite pronounced. The mean size of particles (effective radius) in each mode was determined as

$$\overline{r_{\text{eff}}} = \int_{r_1}^{r_2} \pi r^3 n(r) \mathrm{d}r \Big/ \int_{r_1}^{r_2} \pi r^2 n(r) \mathrm{d}r. \quad (3)$$

Discussion

The averaged data on the spectral variations of AOD in typical Atlantic regions shown in Fig. 2 clearly demonstrate that in different regions they are significantly different both in the value and in the spectrum shape.

The heating of air masses over the continent in the mid-latitudes is not so significant as in the tropics. Here the main mechanism of regular mixing of the continental aerosol with the maritime one is the breeze circulation, which favors the moderate enrichment of

the latter with the fine fraction in a narrow zone (tens of kilometers) along the shoreline. At the same time, under the effect of the particular type of the cyclonic activity of air masses near the continents (as a result of sporadic outflows of the continental aerosol to the maritime atmosphere), the characteristic (in the extent and in the rate of change) trend of AOD variation is observed. Therefore, the NC zone (see Fig. 1) extending by 100 to 200 km from the continent into the ocean and up to 1000 km along the shoreline was separated out[1] in the ocean nearby the continent (in this case, North America). Curve *1* was obtained by averaging the data of optical measurements in this region.

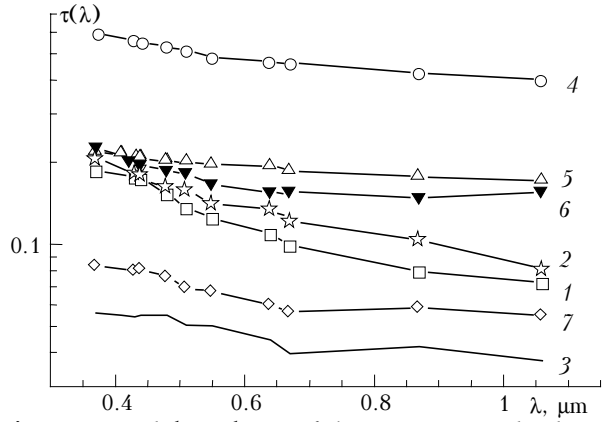

Fig. 2. Spectral dependences of the AOD in typical Atlantic regions: (*1*) near the continent (NC); (*2*) open ocean, moderate (OO) (mod); (*3*) open ocean, background (OO) (b); (*4*) Sea of Darkness (SD); (*5*) trade-wind zone (TW); (*6*) Canary Islands (CI); (*7*) equatorial zone (EZ).

In the central oceanic zone (OO) two states were revealed: moderate turbidity (OO) (mod) shown by curve *2* and minimum turbidity (OO) (b) shown by curve *3*, which can be considered as a characteristic of the background level of AOD in the open ocean. To obtain the latter, the cases of increased aerosol turbidity due to intrusion of continental air masses were excluded from the array of data for this zone. This separation allows us to consider the variations of the aerosol particle number density for different fractions in the zones relative to some common minimum level and to find the features in the size spectrum of aerosol particles generated mostly by the ocean surface.

In the tropical part of the Atlantic, the part of the trade-wind zone adjacent to the African continent and subject to the strongest impact of dust outflows from the Sahara desert was considered separately (curve *4*). The area of moderate and weak effect of dust outflows (curve *5*) is considered as a trade-wind (TW) zone. And, finally, to divide the area of the transverse effect, two more zones are considered: Canary Islands, where the role of dust outflows from the Sahara desert, though being weak, is still considerable, (curve *6*) and the equatorial zone (curve *7*).

Consider first the features of the aerosol microstructure in the mid-latitudes of the Atlantic (Fig. 3). The estimates of the integral parameters for the distributions $s_c(r)$ depicted in Fig. 3 are summarized in Table 1, where V_{ac}, V_{md}, V_{cd} are the integral volumes of particles of the accumulative $r < 0.4$ μm, medium-disperse $0.4 < r < 1.2$ μm, and coarse-disperse $r > 1.2$ μm fractions depending on the particular realization in different zones; V_{mc} is the total volume of the medium and coarse fractions, V is the total volume of all the fractions. The last three columns of Table 1 give the estimated average particle size calculated by Eq. (3) for the three fractions.

The comparison of curves *1–3* in Fig. 3 with the distribution $s_c(r)$ for the continental conditions (curve *4*) reveals their difference at $r > 1$ μm. Under the continental conditions, coarse particles are optically invisible in this example, because their contribution to τ(λ) in the wavelength region used is negligibly small.

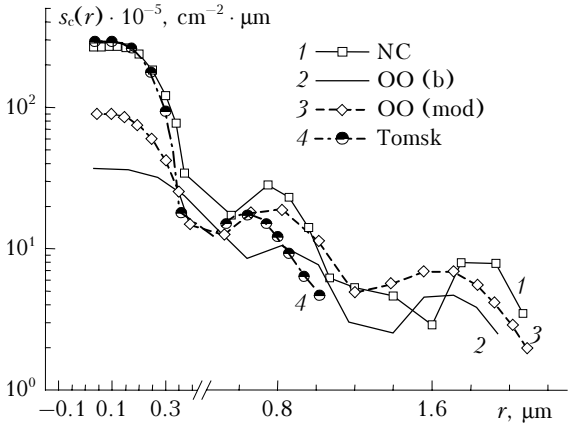

Fig. 3. Curves *1–3* are the distributions $s_c(r)$ retrieved from the data on the spectral variations of AOD (Fig. 2) for the mid-latitudes of the Atlantic; curve *4* is the same but retrieved from the data of optical measurements in Tomsk (June 2000).

It can be seen from Fig. 3 that the size spectra of maritime aerosols in the OO (mod) zone and in the NC zone are similar: the effective radius of the accumulative fraction is ~ 0.16 μm, and that of the medium fraction is somewhat larger than 0.8 μm (Table 1).

Table 1. Integral parameters of the particle size distribution in the mid-latitudes of the Atlantic

Zone	Total particle volume, μm$^3 \cdot$ cm^{-2}					Effective radius, μm		
	V	V_{ac}	V_{mc}	V_{md}	V_{cd}	$\overline{r_{ac}}$	$\overline{r_{md}}$	$\overline{r_{cd}}$
NC	$3.87 \cdot 10^3$	$1.56 \cdot 10^3$	$2.31 \cdot 10^3$	$1.46 \cdot 10^3$	$8.50 \cdot 10^2$	0.16	0.87	1.82
OO (mod)	$2.66 \cdot 10^3$	$5.50 \cdot 10^2$	$2.11 \cdot 10^3$	$1.14 \cdot 10^3$	$9.70 \cdot 10^2$	0.17	0.81	1.65
OO (b)	$1.77 \cdot 10^3$	$4.50 \cdot 10^2$	$1.32 \cdot 10^3$	$6.20 \cdot 10^2$	$3.87 \cdot 10^2$	0.22	0.82	1.65

The main difference between the $s_c(r)$ spectra in the NC and OO zones is different content of aerosol particles with $r < 0.4$ μm. The value of the V_{ac} decreases roughly threefold, and the average particle size partly increases. If we assume that the background level corresponds to purely maritime conditions of particle generation, then the mean contribution of the continental aerosol to the accumulative fraction of aerosol over the ocean amounts to about 70% in the NC zones and 20% in the OO zones.

The situation with particle characteristics in the size range from 0.4 to 2.0 μm is different. The volume contents V_{md} and V_{mc} in the NC and OO zones are roughly equal, but almost twice as high as the background level. There are also some differences as compared to the continent: the effective radius of the medium fraction over the ocean has higher values: from 0.8 to 0.9 μm (for the continent $\overline{r_{md}} < 0.7$ μm). Note also that in the optical manifestation the major role belongs to the accumulative and the medium fractions, while the contribution of coarse fraction only slightly exceeds the error of AOD determination. For a reliable determination of the parameters of this fraction, the information about the spectral dependence of AOD at the wavelengths longer than 1 μm is needed.

The distribution function of the maritime aerosol in the background situations is characterized not only by the minimum values, but also by the weak manifestation of individual fractions. Thus, the decisive role in the spatiotemporal variations of the aerosol in the regions adjacent to Europe and North America belongs to the processes of outflow of the accumulative aerosol fraction from the continents.

In the equatorial zone, the distribution $s_c(r)$ (curve *4* in Fig. 4) is more typical for the continent (curve *4* in Fig. 3) than for the ocean. In the size spectrum there are almost no large particles ($r > 1$ μm), the mode of the medium fraction lies nearby $r = 0.6$ μm. The formation of the accumulative fraction in this zone is affected by the relative closeness to the continent, namely, equatorial Africa. Correspondingly, the quantitative characteristics of the accumulative aerosol fraction (Table 2), such as the total volume of particles, the mean radius, and others, are close to those in the NC zone. At the same time, the above mentioned features of the particle size spectrum over the ocean in EZ likely reflect other conditions of generation and sink of maritime aerosol at these latitudes: gentle wind, frequent precipitations, high humidity and temperature of the air.

It is well known that tropical areas are characterized by the increased atmospheric turbidity due to the trade-wind transport of dust aerosol from Sahara. It can be seen from Fig. 4 and Table 2 that in the Sea of Darkness adjacent to Africa the differential and integral characteristics in the entire size spectrum have the values of an order of magnitude higher than in the background situation.

Even in the TW zone remote from the continent by more than 2000 km and in the area of the Canaries (on the periphery of the trade-wind transport) the aerosol content is higher than in any other region and exceeds the background level by several times.

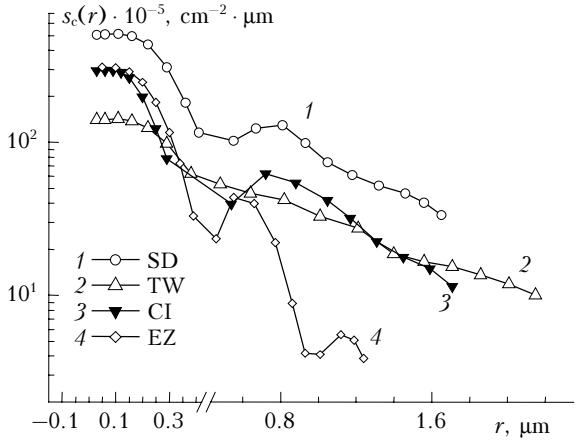

Fig. 4. Distributions $s_c(r)$ retrieved from the data on spectral variations of AOD (Fig. 2) for the tropical latitudes of the Atlantic.

Table 2. Integral parameters of the particle size distribution in the tropical latitudes

Zone		Total particle volume, μm^3 · cm^{-2}			Effective radius, μm	
		V	V_{ac}	V_{mc}	$\overline{r_{ac}}$	$\overline{r_{mc}}$
SD	1	$1.68 \cdot 10^4$	$3.8 \cdot 10^3$	$1.30 \cdot 10^4$	0.18	0.90
	2	$1.97 \cdot 10^4$	$3.8 \cdot 10^3$	$1.59 \cdot 10^4$	0.18	1.10
TW	1	$7.80 \cdot 10^3$	$1.2 \cdot 10^3$	$6.6 \cdot 10^3$	0.19	1.18
	2	$8.80 \cdot 10^3$	$1.08 \cdot 10^3$	$6.7 \cdot 10^3$	0.18	1.19
CI	1	$7.1 \cdot 10^3$	$1.2 \cdot 10^3$	$5.9 \cdot 10^3$	0.14	0.95
	2	$7.5 \cdot 10^3$	$1.3 \cdot 10^3$	$6.3 \cdot 10^3$	0.14	1.07
EZ	1	$3.2 \cdot 10^3$	$1.6 \cdot 10^3$	$1.6 \cdot 10^3$	0.16	0.62

Note. The numbers 1 and 2 in the second column denote the versions of the *a priori* choice of the refractive index for two fractions, namely, (1) $n_{ac} = n_{mc} = 1.45$; (2) $n_{ac} = 1.45$; $n_{mc} = 1.5$.

If in the mid-latitudes the contribution of the accumulative fraction of the continental aerosol is about tens percent, then in the tropics the effect of the aerosol outflows from the continent is noticeable through the entire size range analyzed; in fact, these outflows determine the disperse structure of the aerosol. At the same time, as we go on from the Sea of Darkness to the trade-wind zone, the volume content in the size range $r > 0.4$ μm V_{mc} halves, which is indicative of the weaker effect of the dust aerosol from the Sahara desert with the increasing distance from the African continent.

To the north from the Sea of Darkness, the mixed zone of the Canary Islands was separated out. Depending on the seasonal conditions of air mass circulation, this zone is alternatively affected by the outflows of the fine and coarse aerosol from the two continents. From the northeastern sector, it is enriched with the accumulative aerosol from Europe, and from the eastern and southeastern direction it intakes the continental aerosol from Africa. These effects of the continents manifest themselves in the disperse composition of aerosol: the integral parameters

of the accumulative fraction (the total volume and the mean radius) are close to those of the NC zone (see Tables 1 and 2), and the parameters of the microstructure of the larger particles ($r > 0.4$ μm) agree with the data from the trade-wind zone.

The characteristic feature of the tropical zone is the smoothed character of the distribution functions $s_c(r)$, because of which it becomes senseless to distinguish between the medium and the coarse fractions. Therefore, Table 2 presents only the total data on these fractions.

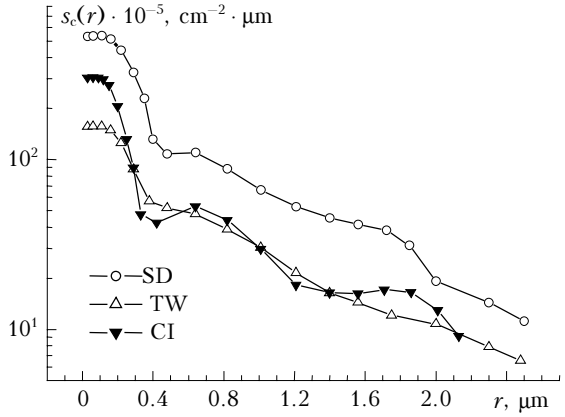

Fig. 5. Distributions $s_c(r)$ retrieved for the tropical zones at the other value of the refractive index ($n_{ac} = 1.45$, $n_{mc} = 1.50$; $\chi = 0.005$).

Taking into account that the atmosphere of the trade-wind zone contains a lot of mineral aerosol, it could be expected that the refractive index of coarse particles ($r > 0.4$ μm) in this zone differ from that in other zones. The effect of the choice of the refractive index on the accuracy of determination of the aerosol microstructure characteristics can be evaluated based on the data given in Table 2, where the rows 1 and 2 present two versions of calculation of the integral parameters at different n_{mc}. The calculated results have shown that the difference in the particle volumes and the effective radii is at the level ~20%. The effect of the refractive index on the differential distribution function can be seen from the comparison of Figs. 4 and 5.

Some changes actually occur in the function $s_c(r)$, but the features of the disperse aerosol composition and the main difference between the zones keep true. Thus, on the whole, the analysis carried out gives the correct idea of the characteristic features of the aerosol composition in the typical zones of the Atlantic.

In conclusion, consider the roles that different aerosol fractions play in the formation of the aerosol optical depth of the maritime atmosphere. It can be seen from Fig. 6 that in the mid-latitudinal zones (OO, NC) and the equatorial zone the optical contribution of the accumulative fraction to AOD decreases monotonically with the increasing wavelength, but still remains prevailing all over the visible spectral region. In the IR region, the roles of the two fractions are roughly identical. In the TW zone, the optical contribution of the coarse particles prevails in the entire spectral region.

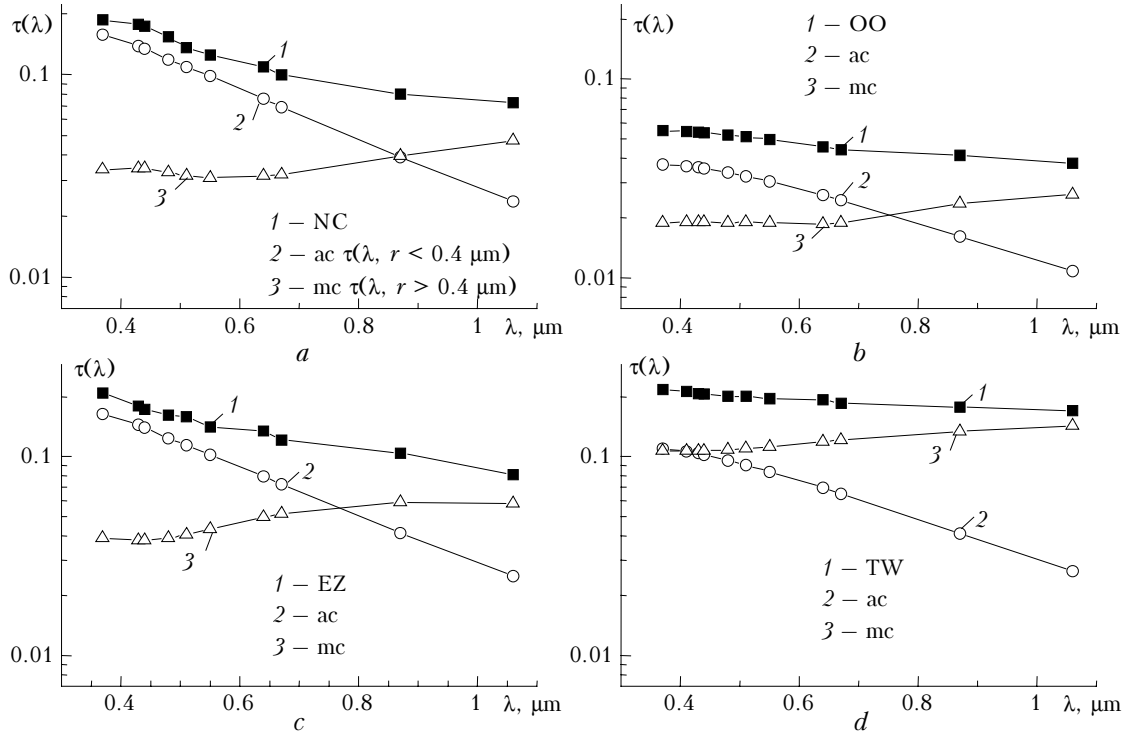

Fig. 6. Spectral variation of the relative optical contribution of different fractions in various zones of the Atlantic: (*a*) near the continent (NC); (*b*) open ocean (OO); (*c*) equatorial zone; (*d*) trade-wind zone.

Conclusions

This analysis is the first stage in investigation of the disperse composition of the maritime aerosol, and the microstructure was retrieved only from the mean spectral dependences of AOD for the earlier separated zones of the Atlantic. Therefore, the particular attention was paid to the search of the most characteristic features in the formation of the disperse composition of the maritime aerosols.

The results obtained demonstrate that the principle of zoning of the maritime atmosphere based on the zonal distribution of the climate and meteorological characteristics is inapplicable to the aerosol component. The main factors determining the regional features of AOD formation over the ocean are the processes regulating the depth and direction of the outflows of the air masses enriched with aerosol particles of continental origin.

In the tropics, in the trade-wind zone and over the Sea of Darkness, the dust inflows from the African continent cover the whole particle size range optically significant for AOD measurements in the wavelength region from 0.37 to 1.06 μm. In fact, they determine the value and nearly neutral spectral profile of $\tau(\lambda)$.

In the mid-latitudes of the Atlantic, the outflows of the continental aerosol determine the content and the mean size of particles of the accumulative fraction. For larger particles, the close values of the total volumes and the mass concentration in the states of the moderate (OO) (mod) and the minimum (OO) (b) turbidity can be explained by the more important role of the conditions of generation and sink of the medium and coarse aerosol than that of the continental impact.

In general, the specificity of formation of the aerosol component in every zone is seen in variation of the value and characteristic percentage of the three aerosol fractions: accumulative ($r < 0.4$ μm), medium-size ($0.4 < r < 1.2$ μm), and the coarse ($r > 1.2$ μm) one.

Acknowledgments

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