# Interpretation of the anomalous spectral dependence of aerosol optical depth of the atmosphere. Part 3. Dynamics of the aerosol disperse composition

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Some peculiarities of the formation of the spectral behavior of the aerosol optical depth are discussed. It is shown that the observed variety of spectral behaviors  $\tau(\lambda)$  is formed due to variations in the optical effect of, at least, three aerosol fractions: accumulative, intermediate dispersed (ID), and specific coarse. Each of the aforementioned fractions has its peculiar rhythm of development and response to the diurnal variations of meteorological quantities. According to data obtained, optical manifestation of the accumulative fraction reacts not only on the change of temperature and relative humidity, but also its behavior agrees with the columnar water vapor of the atmosphere. It is revealed that the narrow mode of intermediate dispersed fraction is characteristic of not only clean arctic air, but it is also observed in the columnar mean size spectrum, under ordinary conditions. Diurnal variations of the ID fraction in the atmospheric column are an evidence of the presence of maximum near noon. Correlated dynamics of the variations of second and third modes shows a stable diurnal circulation of the ID-fraction particles inside the atmospheric column and a change of their size.

#### Introduction

The event of anomalous spectral behavior of the aerosol optical thickness (AOT) of the atmosphere  $\tau(\lambda)$ observed in Tomsk at intrusion of clean Arctic air was discussed in Refs. 1 and 2. It was shown, based on microphysical modeling and solving the inverse problem, that the anomalous dependence is caused by the peculiarities of aerosol disperse composition, namely, by the deficiency of the particles of accumulative and coarse fractions and the presence of narrow mode of intermediate dispersion (ID) fraction with the modal radius of  $\sim 0.5 \,\mu\text{m}$ . The ID-fraction particles determined non-typical behavior of  $\tau(\lambda)$  in the form of diffuse maximum in the red wavelength range due to their increased optical significance. The noted dependence is typical for Antarctica and is characteristic of the background global aerosol of the atmospheric column.<sup>3,4</sup> The value of the modal radius of particles of 0.5 µm was obtained from inverting the mean spectra  $\tau(\lambda)$  of the Antarctic atmosphere.<sup>2</sup> Analogous data ( $r_{\rm m} \sim 0.5 \ \mu {\rm m}$ ) for Antarctica are presented in Ref. 5 based on solving the inverse problem for aureole scattering phase functions.

Stability of Tomsk anomalous spectral dependence  $\tau(\lambda)$  and corresponding aerosol disperse structure was observed<sup>1</sup> during several days. However, as subsequent analysis has shown, this conclusion is true because of the general tendency of day-to-day variations of the haze microstructure and the optical data obtained in different days but approximately at the same time of a day, at noon, in our case. More detailed analysis of diurnal variability of the haze microstructure has revealed quite complicated dynamics of the proportions

between the content of accumulative and ID fractions. This fact predetermines the content of this paper.

Hourly mean spectra of AOT of the atmosphere acquired since July 23 until July 29, 1997 were used for inverting the optical data. This period included both an anomalous situation and ordinary conditions. Besides, the data were incorporated in the analysis for a few days with weak clouds, like on July 1999 with detailed (in time) measurements of  $\tau(\lambda)$  and meteorological characteristics.

# 1. Day-to-day variations

The technique for solving the inverse problem briefly described in Ref. 2 was applied to interpretation of the spectral measurements of AOT. The sought distributions of  $s_c(r)$  was found from solution of the system of equations of the form

$$\int_{R_1}^{R_2} k_{\varepsilon}(r, \lambda_i) s_{c}(r) dr = \tau(\lambda_i), i = 1, 2, ..., n, \quad (1)$$

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 extinction efficiency factor for radiation that depends on the complex refractive index of particles  $m = n - i\chi$ ;  $R_1$  and  $R_2$  are the boundaries of the sought function  $s_c(r)$ . It was assumed in inverting that the real part of the complex refractive index n = 1.45 and the imaginary part  $\chi = 0.005$ .

The examples of thus reconstructed particle size distributions are presented in the form of the right-hand boundary  $R_2$ , the relationship for estimating this value was taken from Ref. 2. The obtained estimate

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 $R_2 \cong 1.0 \ \mu\text{m}$  works in the majority of cases and only sometimes, it reaches 1.4  $\mu\text{m}$ .

Spectral dependences  $\tau(\lambda)$  measured at 10 wavelengths in the range from 0.35 to 1.06 µm were used for inversion. The effect of measurement error in AOT on the accuracy of determination of the aerosol microstructure parameters was studied in the preliminary numerical experiments. Some results of these investigations are shown in Fig. 1. To estimate the effect of the error, the daily mean dependence  $\tau(\lambda)$  obtained under stable atmospheric conditions was disturbed by oscillating error following the relationship

$$\tau^*(\lambda_i) = \tau(\lambda_i) + (-1)^{i+1} \delta_{\tau}(\lambda_i).$$
(2)

The error in determining AOT  $\delta_{\tau}(\lambda_i)$ , according to estimates from Ref. 1, is not uniformly distributed over the wavelength range, varying in the limits 0.005 to 0.01 (Fig. 1). The maximum relative error lies at the wavelength of 1.06 µm and is about 30% of the measured value. Corresponding errors in determining the total geometric cross section *S* and the volume *V* of particles (entire submicron aerosol), and analogous parameters of the accumulative fraction  $S_1$  and  $V_1$  are at the level of 3–4%.



Fig. 1. Effect of the errors in optical measurements on the accuracy of solution of the inverse problem.

The error in determining the total volume of IDfraction particles  $V_2$  is the largest; in this case, it is  $\pm 12\%$ . On the whole, numerical estimates show that random errors of the real experiment are not the principal distortions of the results of optical data inversion.

The set of anomalous and ordinary spectral dependences of AOT of the atmosphere was obtained from observations in Tomsk in different years (Fig. 2a). The data of one month, July, were selected in order to avoid possible seasonal variations. It is seen in the results of inversion (Fig. 2b) that the bimodal shape of the particle size distributions estimated in the atmospheric column is observed under both anomalous

and ordinary conditions. Besides, the results obtained show relatively independent dynamics of the formation of the particle size spectrum in the aforementioned size ranges.



Fig. 2. Examples of daily mean spectral dependences of AOT of the atmosphere and the results of their inversion.

Noticeable differences the differential in distribution functions  $s_c(r)$  are seen in the position of the maximum of the second mode. Under ordinary conditions related to continental air masses the maximum lies in the particle size range 0.65 to  $0.7 \mu m$ , and under conditions of anomalous transparency ID fraction contains smaller particles. Its modal radius in this case lies in the range 0.55 to 0.58 µm. Analysis of the measured anomalous spectral dependences of AOT obtained in different regions (Antarctica, Arctic (SP-22),<sup>3</sup> village Sosnovo, Leningrad region,<sup>6</sup> Ryazan,<sup>7</sup> Tomsk,<sup>1</sup> Abastumani,<sup>8</sup> and the results of their  $interpretation^1$  show that the more spectral AOT approaches to the background state, the less accumulative particle fraction is pronounced and the more noticeable the modal radius of ID fraction is displaced to the range  $r \leq 0.6 \,\mu\text{m}$ .

It is seen in Fig. 2 that there is the well pronounced boundary between accumulative and

intermediately dispersed fractions, the position of which varies in the range 0.3 to 0.45  $\mu$ m as atmospheric conditions change. The boundary between these fractions mainly depends on the right boundary of accumulative mode, which can move depending on the intensity of generation of secondary aerosols. The integral estimates of microstructure parameters of each fraction of particles were determined taking into account this boundary. The estimates of the integral parameters were also determined using approximation of the size spectrum of each fraction by an analytical dependence (4-parameter gamma-distribution). Differences in the estimates are 10–15%.

The mean radius of particles of the accumulative mode determined by the relationship  $% \left( {{{\left( {{{{{\bf{n}}}} \right)}}_{{{\bf{n}}}}}} \right)$ 

$$r_{\text{mean}} = \int_{r_1}^{r_2} \pi r^3 n(r) \, \mathrm{d}r \, / \, \int_{r_1}^{r_2} \pi r^2 n(r) \, \mathrm{d}r \qquad (3)$$

lies in the range of 0.14 to 0.17  $\mu$ m for all realizations  $s_{\rm c}(r)$  shown in Fig. 2.

Some peculiarities in the transformation of aerosol microstructure since July 23 until July 29, 1997 are shown in Fig. 3 as compared with the dynamics of nearground temperature T and columnar water vapor W density. Let us remind that the above noted time interval covers full cycle of the process of the change of the following synoptic conditions:

a) on July 24 continental mid-latitude air was replaced by the Arctic air mass with a drastic fall of the air temperature and water content;

b) anomalous spectral behavior of AOT was observed since July 24 until July 26 under conditions of decreased temperature and humidity;

c) on July 27 Arctic air mass was replaced by continental mid-latitude one with the corresponding increase of the air temperature and humidity.

The change of synoptic conditions was observed in the dynamics of aerosol microstructure as follows. As Arctic air mass had invaded, the total volume of the accumulative fraction of particles  $V_1$  decreased by several times and returned to previous values only in two days after continental mid-latitude air mass had come. Day-to-day dynamics of the total volume of the ID fraction of particles at the same time was insignificant. Some increase of daily mean values  $V_2$  on July 24 and July 25 can be related to the errors in measuring and solving the inverse problem, as well as to small quantity of data obtained in the evening due to bad weather conditions (cloudiness).

It is interesting to estimate optical contribution of two fractions to AOT in the extreme cases (July 24 and July 28) at passing from anomalous transparency to ordinary one. To do this, the values  $\tau(r)$  at the wavelength of 0.55 µm were calculated as functions of the upper limit of integration in equation (1). It is seen in Fig. 4 that under typical conditions the accumulative aerosol fraction determines 80% of daily mean values of AOT. On the contrary, optical contribution of intermediately dispersed particles, which are about 70%, prevails under anomalous conditions.



Fig. 3. Dynamics of daily mean total volumes of particles of the aerosol fractions, column density of water vapor, and nearground temperature.



Fig. 4. Relative optical contributions of aerosol of different size under anomalous (July 24, 1997) and ordinary conditions (July 28, 1997) assessed from optical measurements of AOT in Tomsk.

## 2. Diurnal dynamics

Diurnal dynamics of the spectral behavior of AOT (Fig. 5) is complicated and does not enable us to unambiguously interpret even the limited set of atmospheric conditions. The results of solving the inverse problem show that such a behavior  $\tau(\lambda)$  is significantly caused by the fact that the processes determining the mechanism and rhythm of diurnal variability of the content of particles of accumulative and intermediate fractions in the atmospheric column are noticeably different. This predetermines different reaction of the particles of accumulative and intermediate fractions on the change of meteorological conditions.

It is seen from the experimental data shown in Fig. 5 that local deviations from the Angström dependence are characteristic of many realizations of  $\tau(\lambda)$  under ordinary (not anomalous) conditions. Numerical analysis shows that one cannot ignore these deviations, because they are caused by real aerosol size spectra, which are more complicated than simple analytical models of the Junge type or one-modal lognormal law. It should be noted that the attempts to use the model stylizations (which reflect only general tendency in the change of the disperse structure of atmospheric aerosols) are not always justified for describing the specific spectra.

Besides, the data in Fig. 5 show not only different rate of the change of  $\tau(\lambda)$  at different wavelengths, but mutually contrary tendencies, that is evidence of the presence of relatively independent mechanism of the formation of aerosol subfractions in the size ranges 0.08–0.35 and 0.45–0.9 µm. Authors emphasize that the revealed peculiarity is characteristic of the aerosol size spectrum averaged over the atmospheric column. The aerosol size spectrum in the analyzed range in the near-ground layer is smoothed due to intense influence of a great number of the sources of submicron aerosol.

The enhanced values of AOT in the red spectral range relative to the values obtained by extrapolation

of the optical characteristics measured in the shortwave range by the Angström formula enable us to draw a conclusion on the presence of intermediate-disperse fraction, that the solution of the inverse problem reveals.

On some days, the volume of accumulative fraction  $V_1$  (Fig. 6a) follows the morning decrease of near-ground relative humidity (Fig. 6c). The mean radius  $r_{m1}$  of the accumulative fraction simultaneously decreases (Fig. 6b), that is evidence of the loss of moisture by aerosol. At the same time, the increase of the volume of the particles of accumulative fraction since morning is observed in some cases. The observed variety of the tendencies of diurnal variations of  $V_1$ quite stably correlates with the change of the water vapor column (see Fig. 6a). Some deviations from the coordinated dynamics of these values are related, as a rule, to the change of the state of the atmosphere, for example, to the appearance of visually observed haze on July 23 (1 p.m.) and passing of a cloud with rain of short duration (2 p.m.).

Let us emphasize that we say only about the stable correlation of variations of the discussed parameters, which was observed in many cases of diurnal and daily dynamics (see Fig. 3, 6a, and b) and, as a rule, inside the same air mass. It is unlikely that the observed correlation at an arbitrary data set is the casual coincidence of circumstances. It is more likely, that this fact is evidence of the presence of a mechanism making the accumulative fraction dependent on the water vapor content. It is well known that clean arctic air, at least in summer, is characterized by extremely low aerosol content.<sup>12</sup> The deficiency of secondary aerosol is significantly caused by exhausted composition of aerosol producing gases and water vapor. The motion of arctic air deep in the continent is accompanied by enrichment of the composition, increase of temperature, humidity and insolation, i.e., in addition to the process of air mixing, favorable conditions are formed for additional development of the accumulative fraction to the normal level.



Fig. 5. Examples of diurnal variability of the spectral dependence of AOT under different atmospheric conditions in arctic (July 24, 1997) and continental (July 28, 1997, July 6, 1999) air masses.



Fig. 6. Diurnal behavior of the volume  $V_1$ , mean radius  $r_{m1}$  of the accumulative-fraction particles, column density of water vapor of the atmosphere W, and near-ground relative humidity of the air RH.

Of coarse, few examples are insufficient for making a statement on the aforementioned correlation. Additional investigations are necessary for revealing specific mechanism of the influence that the moisture content of an air mass can produce on the content of the accumulative-fraction aerosol in it. Let us also note that the presented hypothesis does not contradict the earlier conclusion on the ambiguity of the dependence of AOT on the atmospheric water vapor column density.<sup>13</sup> The kinetics of the formation of disperse composition of the accumulative fraction, which was partially discussed earlier,<sup>2</sup> proceeds on the background of various interactions among meteorological factors. So the correlation between the variations of  $V_1$  and W noticeably decreases when analyzing the data obtained during a significantly long time interval.

The results presented only confirm the conclusion that the prehistory of the development of the air mass, trajectory of its regional motion, and interaction with other air masses, plays an important role in the formation of the size spectrum of the accumulative fraction. Some features, which were hidden or ambiguous in previous analysis of the mechanism of

fraction. Some features, which were hidden or ambiguous in previous analysis of the mechanism of formation of the integral optical characteristic  $\tau(\lambda)$ ,<sup>13</sup> may be revealed in analyzing correlations between separate aerosol fractions. In other words, analysis of the characteristics of disperse composition and the tendencies in the relative variability of separate fractions (but not correlation with  $\tau(\lambda)$ ) opens additional approaches to understanding specific mechanisms of aerosol transformation under the effect of varying environment.

Integral parameters of the size distribution of the intermediate disperse fraction  $(S_2, V_2)$  for the considered realizations of  $\tau(\lambda)$  (including the events of anomalous transparency) vary analogously to the diurnal behavior of the total solar radiation Q (Fig. 7). The volume and geometric cross section of the particles of this fraction increase by noon, reach maximum at 1-2 p.m., and then monotonically decrease. The portion of  $V_2$  in the total volume of submicron aerosol, in the case of ordinary spectral behavior of AOT, increases from 25% in the morning to 40-45% at noon and decreases to 10% at night. Diurnal behavior of the atmospheric transmission in the visible wavelength range is formed in this case under prevalent effect of the accumulative fraction. The optical contribution coming from the intermediate fraction at the wavelength of  $0.55~\mu m$  does not exceed 10% in the morning (9 a.m.) and in the evening (8 p.m.), while increasing up to 25% by noon.

The ratio of the volumes of accumulative and ID fraction under conditions of anomalous transparency of the atmosphere essentially changes during a day. In this case, one can observe the principal tendencies in the diurnal behavior of the reconstructed distributions  $v_c(r)$ in Fig. 8. In the morning (9-10 a.m.) the total volumes of particles of accumulative and ID fractions are more close values, and their optical contribution at the wavelength of 0.55  $\mu$ m is about 50%. Such a ratio of the fractions makes the spectral behavior  $\tau(\lambda)$  to be nearly the ordinary one (see Fig. 5). Resulting from an essential increase of the volume  $V_2$  against the background of some increase in  $V_1$  by noon, the optical contribution of the intermediate-fraction particles dominating that leads to anomalous becomes transparency with enhanced values of AOT in the red spectral range (see Fig. 5a).

One can see another one peculiarity in the diurnal behavior of the parameters of the intermediate fraction observed under conditions of cloudless atmosphere on July 28, 1997 (Fig. 9*a*). Simultaneously with the morning increase of  $V_2$ , the modal radius of ID fraction successively moves to smaller values. Analogous tendency in the morning variation of the mean radius of

the particles of ID fraction  $(r_{m2})$  covers all the period since July 24 until July 29, 1997, including the event of anomalous transparency (Fig. 9b). Another type of the temporal behavior of  $r_{m2}$  was observed on July 6, 1999. The increase of the mean particle size in the morning in this case shows, evidently, the additional (relative to the period from July 24 until July 29, 1997) generation of the ID-fraction particles, the intensity of which decreases as the rate of temperature growth decreases (Fig. 9c). One can suggest the preliminary hypothesis that the additional source in this case could be destruction of coarse particles. The reason for such supposition is the enhanced content of particles with the radii ranging from 1 to  $1.5 \ \mu m$  observed in the morning on July 6, 1999. Let us also note that the total volume of particles decreased following the increase of  $r_{\rm m2}$  at the temperature increase that took place by 3 p.m.



**Fig. 7.** Diurnal behavior of the volume of particles of intermediate fraction  $(V_2)$  and the total solar radiation.



**Fig. 8.** diurnal variation of the density of the volume size distribution of haze particles v(r) (July 24, 1997) under conditions of anomalous transparency of the atmosphere.

Numerical estimates show that the total optical contribution of coarse aerosol in this case is much lower than the measurement error even at the wavelengths of 0.87 and 1.06  $\mu$ m. However, the contribution of these particles at their destruction can be observed in the red spectral range. The results of experiments in a closed volume using a photoelectric counter and a filter trap<sup>9</sup> evidence of the possibility of the effect of the mechanism of disaggregation of coarse particles at certain temperatures of the air followed by the appearance of additional number of small particles. However, to draw more valid conclusions on the change of the size spectrum of coarse fraction, it is necessary to expand the spectral range of AOT measurements to IR range for solving the inverse problem for the real atmosphere.

One can trace some signs of the disaggregation process in our results shown in Fig. 10.

Spectral dynamics of v(r) was obtained from the measurements data acquired on June 23, 1999. First, the fact attracts our attention that the column content of coarse aerosol fraction successively decreases during the day, and by the evening these particles practically disappear from the spectrum from the standpoint of their contribution into the spectral dependence of AOT. Second, unusual increase of the content of particles of the size from 0.4 to 0.75 µm is observed during the same day in comparison with other episodes of measurements, that is approximately 2-2.5 times less than the characteristic size of the coarse-fraction particles. The size range that was actually filled during a day is intermediate between the accumulative and intermediate, or haze fractions. The noted peculiarity in deformation of the size spectrum was observed only once. Strict separation between the two fractions kept in the majority of other episodes.

The most probable cause of the revealed peculiarity could be activation of an additional local source of aerosols of a certain size, for example, smoke aerosols. The results of optical measurements and theoretical estimates<sup>10,11</sup> confirm this fact. In particular, according to theoretical estimates,<sup>11</sup> high number density of fine aerosol with  $r \sim 0.01$  to  $0.05 \ \mu m$ , which is effectively transformed to the aerosol fraction of the characteristic size from 0.25 to 0.35 µm is reached at thermal sublimation of combustible matters. The noticeable deformations of the size spectrum of accumulative fraction appear at the ordinary level of the content of fine aerosols only in connection with the change of the total water content inside the air mass or relative humidity in the nearground layer.



Fig. 9. An example of diurnal dynamics of the distribution s(r), mean radius of particles of the second mode  $r_{m2}$ , and near-ground temperature T at anomalous and ordinary spectral behavior of the AOT.





**Fig. 10.** Diurnal variation of the density of the volume size distribution of haze particles v(r) reconstructed from the measurement data on AOT on June 23, 1999.

Correlated dynamics of variations of the contents of intermediate and coarse-fraction particles is also seen in the measurement results obtained on July 6, 1999 (Fig. 11*a*). It is easy to see that an essential increase in the ID fraction content is observed by 2 p.m., which is accompanied by practically complete disappearance of the coarse fraction of particles (Fig. 11*b* and Table). At the same time, as near-ground temperature increases and relative humidity decreases, the size spectrum of accumulative fraction becomes narrower, the mean radius decreases (see Fig. 6*b*), and the specific content decreases.

General dynamics of the change of the mean radius of particles of ID fraction is shown in Fig. 9b. It is interesting to note (see Table) that quantitative increase of the volume of the intermediate-fraction particles occurred since 10 a.m. by 2 p.m. is  $\Delta V_2 = 430 \ \mu m^3 / \text{cm}^2$ , that is in a good agreement with the total volume of the third mode of particles observed in the morning and their disappearance from the mean size spectrum after 3 p.m. Diurnal dynamics of the spectrum transformation is an evidence of the opposite change of the contents of second and third modes in the ensemble of aerosol particles.

The column mean spectrum of atmospheric haze particles in the evening has characteristic bimodal shape (Fig. 11c). However, in contrast to the data acquired on June 23, 1999, no filling of the inter-fraction interval was observed in this case, as in the majority of other measurements. In other words, the episodes of extension of the size spectrum of accumulative fraction and their confluence with the mode of intermediate fraction at low turbidity of the atmosphere are quite rare and are observed at active generation of fine aerosols, for example, from smoke.

Different behavior of two fractions of submicron aerosol in the afternoon attracts our particular attention. In contrast to the accumulative fraction, a well-pronounced decrease in the content of ID fraction is observed by the evening. The causes of a more effective decrease of the content of the intermediatefraction particles are not quite clear. Such a change cannot be explained by sedimentation process because of a small size. Although the efficiency of the Stokes sedimentation of ID particles is a little bit higher than that of the accumulative fraction (~  $2 \cdot 10^{-4}$  cm/s), however, it is very small (~  $3 \cdot 10^{-3}$  cm/s) and it is much less than the effective velocity of dry sedimentation for both fractions (~  $3-9 \cdot 10^{-2}$  cm/s).<sup>14</sup>



Fig. 11. Diurnal variation of the density of the volume size distribution of haze particles v(r) reconstructed from the measurement data on AOT on July 6, 1999.

Table

Time, hour	$V_1 \cdot 10^{-5}, \ {\rm cm}^{-2} \cdot {\rm \mu m}^3$	$V_2 \cdot 10^{-5},\ { m cm}^{-2} \cdot { m \mu m}^3$	$V_3 \cdot 10^{-5},\ { m cm}^{-2} \cdot { m \mu m}^3$
9	0.00737	0.00171	0,0042
10	0.00647	0.00320	0,00319
11	0.00519	0.00415	0,00309
12	0.00496	0.00546	0,00252
13	0.00518	0.00694	0,00245
14	0.00564	0.00752	0,00147
15	0.00707	0.00722	$6 \cdot 10^{-4}$
16	0.00823	0.00644	$10^{-4}$

Two working hypotheses are seen from the set of the revealed peculiarities. According to the first mechanism, the process of convective filling of the boundary layer with the ID-fraction particles in the afternoon reaches the height of formation of cloud structures. When the maximum of the content of IDfraction particles in the boundary layer has been reached, their sink to the area of active cloud formation intensifies in the afternoon. Reaching this height range, the ID-fraction particles are involved into the cloud droplet structure both on the stage of heterogeneous condensation and at subsequent motion inside the cloud layer. Thermal mixing, phoretic forces, and the forces of electromagnetic interaction favor active migration of the ID-fraction particles to cloud droplets. Thus, some ID-fraction particles can be found inside cloud droplets. It is difficult to estimate the rate of losses of aerosol due to inner cloud processes, however, according to Ref. 15, liquid-droplet phase has quite great efficiency of removing insoluble particles ~  $10^{3}$ - $10^{6}$  per cm<sup>3</sup>. At droplet evaporation, these particles are aggregated by surface tension of droplets into unstable structures and can have the initial size 2 to 3 times larger than the characteristic size of intermediate- fraction particles. Possibly, these are observed as the third mode of the size spectrum in some morning realizations of  $\tau(\lambda)$ . Let us emphasize that the afternoon formation of cloud was observed in the majority of the considered days, and measurements were carried out in the space between the clouds.

Another active sink mechanism for the ID-fraction particles can be the process of dry sedimentation on the surface of vegetation (grass, leaves, crowns of trees). Let us add that in this case night cooling and formation of dew also favor their partial aggregation and transition of some ID-fraction particles into an aggregated state with the characteristic size of the aggregation particles characteristic of the third mode. Coming to the convective flow in the aggregated state, these unstable aggregates dry and disintegrate by the noon as temperature increases.

In the case of confirmation of the suggested hypotheses, one can say that particles of the second and third modes are also involved into the long process of secondary inner atmospheric transformation. If the particles of third mode had primarily soil origin, the diurnal dynamics of their content should correspond to the development of convection and maximum of heating the soil. However, the results considered do not agree with that simple scenario of the convective lifting and sedimentation. In other words, the signs of daily circulation of ID-fraction particles are observed in the dynamics of aerosol of the size from 0.4 to 1.7  $\mu$ m.

## Conclusion

Finishing the discussion of the problem as a whole, one can conclude that localization of ID fraction in the column mean disperse composition of the

atmospheric haze is not a rarity characteristics of only clean arctic air. Narrow ID mode is observed in the spectrum of aerosol particles under ordinary conditions, but it becomes priority (determining) in the optical effect under conditions close to the background ones.

Analysis of the dynamical characteristics during the "anomaly" has shown that the disperse structure was not conservative at keeping the dominating role of ID fraction. Each fraction had its own diurnal and daily variations under the effect of the environment. The variety of spectral dependences of AOT (including anomalous one) in the wavelength range from 0.35 to 1.06 µm is formed due to the change of optical effect of the accumulative (the right-hand boundary of the distribution s(r)  $r_2 \approx 0.3-0.4$  µm) and intermediate ( $r_2 \leq 1$  µm) aerosol fractions. Independent rhythms of diurnal dynamics of each of these fractions are evidence of different processes mainly affecting the content of submicron particles of different size in the atmospheric column.

In particular, optical effect of the accumulative fraction agrees with not only the variations of temperature and relative humidity, but also with the column density of water vapor, because these factors form specific conditions of the transition of the vapor of aerosol producing compounds to the fine aerosol phase, from which the optically active aerosols are formed.

At the same time, as the obtained estimates show, diurnal variations of the ID-fraction particles column density and the parameters of their size distribution agree with the change of the conditions of convective lifting and efficiency of their emission into the free atmosphere, to the cloud formation zone, as well as dry sedimentation.

As to the conclusions on the optical contribution of the coarse-fraction particles to AOT, one should consider the obtained estimates as preliminary. However, even these preliminary estimates of the dynamics of the content of the third fraction are quite interesting and deserve attention. It is supposed to study this problem in a more detail with the extension of the spectral range of AOT measurements to the near IR range.

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