# INFLUENCE OF AEROSOLS ON THE SPECTRAL OPTICAL THICKNESS OF THE ATMOSPHERE OVER SOUTHERN PART OF LAKE BAIKAL

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*In this paper we discuss some measurement results of spectral optical thickness of the atmosphere conducted in 1991 in southern part of lake Baikal. The observation sites were located near the points of aerosol sampling. The size spectrum of aerosol particles is well approximated by a sum of three lognormal size distributions. The set of experimental data collected and their theoretical analysis show that the optical depth of the atmosphere over southern part of lake Baikal is mainly determined by the properties of aerosols corresponding to the second mode of size distribution. This result well agrees with the data obtained in other geographical regions.*

Aerosol is an important component of the atmosphere and has a significant impact on the solar radiation balance. Atmospheric aerosols are responsible for visibility in the atmosphere[1,2] and influence the climate.[3] For this reason, the study of optical properties of aerosols is now receiving widespread attention.[4,5] However there are as yet no reliable examinations of the relation between the size spectrum, concentration of aerosols, and spectral optical thickness (SOT).

## OBSERVATION SITE, INSTRUMENTATION, AND MEASUREMENT TECHNIQUE

The SOT measurements were carried out in July 1991 over southern part of lake Baikal.[7] The observation sites were located near the points of aerosol sampling. One of the points was at the meteorological station near the source of the Angara river, and the other one was 5 kilometres to the East at a site of solar telescope. The height of the first point above the level of lake Baikal was 5 m, and the height of the second one was 500 m. The height of the surface of lake Baikal above the sea level was 470 m. The coordinates of the observations points were about 53°N and 104°E.

The SOT measurements were conducted with a Feysner spectral actinometer with a set of interference light filters. The viewing angle was 9°. The temperature correction was about 3% per 10°C. A digital voltmeter F−30 was used as a recorder. Table I lists the basic specifications of the light filters used. The spectral intervals chosen are in fact free from absorption by atmospheric gases, with the exception of ozone in the Chappean band. The correction to it was introduced based on measurements of the total content of ozone in the region of observations carried out with the M−124 ozone meter.

*TABLE I. Spectral characteristics of light filters.*

| Wavelength at maximum, nm | 383 | 425 | 491 | 520 | 556 | 590 | 672 | 760 | 774 | 820 |
|---|---|---|---|---|---|---|---|---|---|---|
| Halfwidth of transmittance, nm | 11 | 10 | 12 | 14 | 12 | 12 | 16 | 17 | 20 | 25 |
| Transmittance at maximum, % | 36 | 37 | 43 | 47 | 37 | 29 | 36 | 32 | 40 | 20 |

The spectral optical thickness was calculated using formula (1), Ref. 6. Formula (2) for calculating optical mass of the atmosphere ($m$) is valid with the error smaller than 1 % for the sun elevation $h > 10°$, Ref. 6. Thus

$$\tau_\lambda = [\log (S_{0\lambda}/S_\lambda) \ / \ m] - \alpha_\lambda \, X - \beta_\lambda \, , \qquad (1)$$

where $S_{0\lambda}$ is the radiation beyond the atmosphere at the wavelength $\lambda$; $S_\lambda$ is the radiation measured at the wavelength $\lambda$; $\alpha_\lambda$ is the absorption coefficient of radiation at wavelength $\lambda$ by ozone; $X$ is the total content of ozone in the atmosphere; and, $\beta_\lambda$ is the optical thickness of the Rayleigh atmosphere; and,

$$m = 796.8 \, \sin(h) \left( \sqrt{1 + (0.05/\sin^2 h)} - 1 \right), \qquad (2)$$

where $h$ is the sun elevation above the horizon.

With an optically stable atmosphere, a plot of $\log S_\lambda$ *vs* $m$ is linear.[6] This allows one to determine the value $S_{0\lambda}$ by extrapolating the experimental dependence to a zero value of optical mass of the atmosphere ($m = 0$). To this end we chose a set of observations on July 12, 1991 since the measurement results of this set spread well on a straight line.

To measure size spectra and number density of aerosol particles with diameter smaller than 0.4 μm we used a lattice diffusion battery TSI − 3040 and a TSI − 3020 counter of condensation centres. Impactors of various types were used for particles larger than 1 μm.

## MEASUREMENT RESULTS AND THEIR ANALYSIS

Figure 1 depicts the SOT measurement results obtained during a day at three wavelengths: 383, 556, and 820 nm.

      

The atmosphere is seen to be the most transparent on July 12, 1991. The strongest extinction of solar radiation was on July 21, 1991.
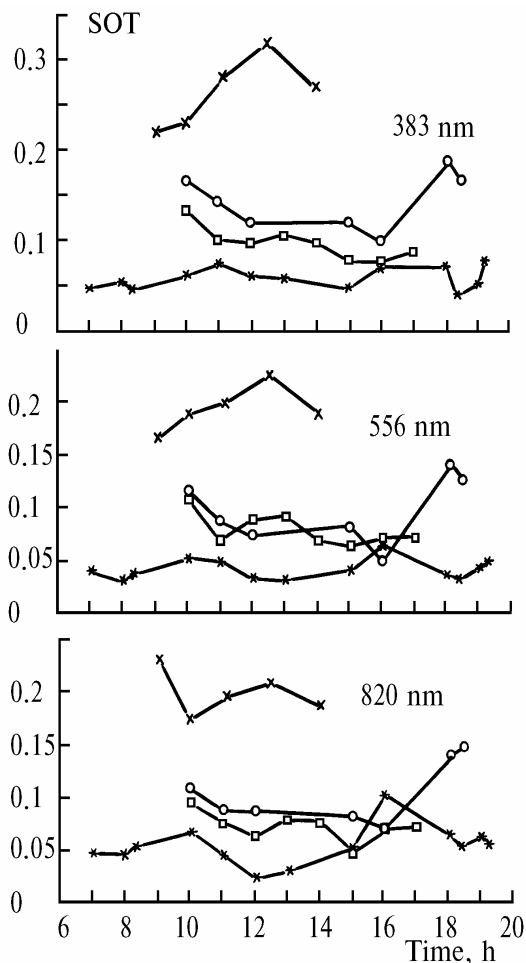


FIG. 1. Diurnal behavior of spectral optical thickness at different wavelength: July 12 (*), July 13 (°), July 21 (×), and July 22 (п).

On the day with steady weather there are two maxima in a diurnal behaviour of SOT. The morning maximum is at about 10—11 and the evening one at 16 —18 h. The diurnal variations of SOT at different wavelengths were usually similar. While on the 12th, 13th, and 22 nd the SOT minimum was observed at 12—14 h, whereas on the day with maximum optical depth (July 21, 1991) the SOT maximum was observed at that time.

To determine the reason of variations of SOT let us consider its change at different wavelengths. Shown in Fig. 2 is a plot of SOT *vs* wavelength on 12, 13, 21, and 22 of July, 1991. The three curves in each figure are related to morning, afternoon, and evening measurements. The spectral behavior of SOT for different days is different but it is approximately the same at different periods of one and the same day. This indicates that the aerosol particle size spectrum, which affects SOT, changes day after day, but remains unchanged during a single day.
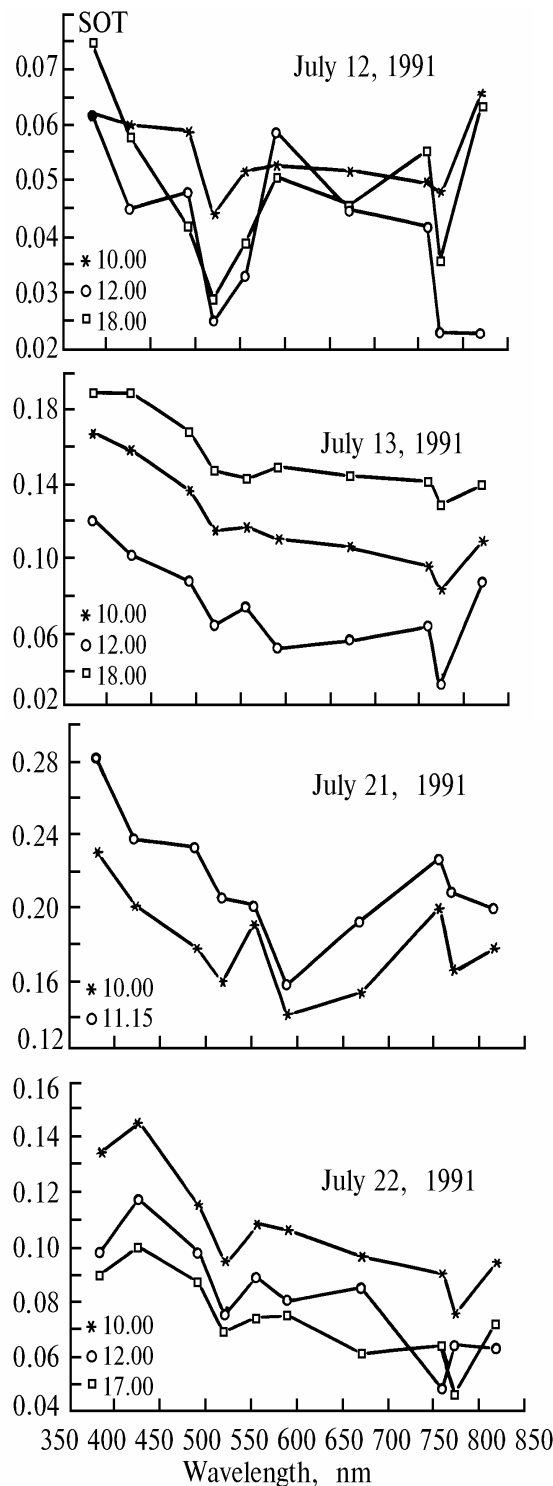


FIG. 2. Spectral optical thickness vs wavelength.

Thus we can infer from these results that during a day the number density of aerosol particles changes substantially whereas the size spectrum affecting SOT is almost constant.

In our measurements SOT did not monotonously decrease with the wavelength increase, in contrast to Ref. 9.

From our standpoint, the reason for this difference may be the averaging of the results over a sufficiently large period of time, which was carries out in Ref. 9. At the same time the nonmonotonic changes of SOT are also described in Ref. 6 and explained by the effect of absorption bands by gaseous admixture on SOT. But this argument is not so convincing since the wavelength dependence of SOT observed in so doing must keep its form. A possible explanation for substantial variations in SOT from experiment to experiment is the variability of size spectrum, concentration, and chemical composition of atmospheric aerosols.

We also measured size spectrum and concentration of aerosols as well as the dynamics of their variations with time. The aerosol particle size spectrum obtained by averaging the measurement results over the entire period of observations[8] is approximated sufficiently well by the sum of three lognormal functions

$$dN/d \log r = \sum_{i=1}^{3} \frac{n_i}{\sqrt{2\pi} \log \sigma_i} \exp \left( -\frac{\log^2(r/R_i)}{2 \log^2 \sigma_i} \right). \quad (3)$$

The parameters of formula (3) resulting from processing the experimental data are listed in Table II.

*TABLE II. Parameters of lognormal approximation of size spectrum of atmospheric aerosol.*

|  | Mode number | | |
|---|---|---|---|
| Parameters | 1st | 2nd | 3rd |
| $n_i$, cm$^{-3}$ | 1100 | 4600 | 0.3 |
| $R_i$, μm | 0.01 | 0.045 | 0.8 |
| $\sigma_i$ | 1.3 | 2.0 | 2.4 |

As seen from examinations of the diurnal behavior of variations in particle number density and size spectrum of submicron fraction of aerosol (the first and second modes), this segment of the size spectrum is related to their photochemical formation and subsequent aging due to coagulation.[8] These results are well confirmed by the experimental data (Figs. 3 and 4). Figure 3 depicts the diurnal behavior of variations in particle number density, and Fig. 4 illustrates a character of transformation of the submicron aerosol particle size spectrum at different time of the day. In particular, in Fig. 3 there is a pronounced maximum of particle number density which was formed some hours later after the sun rise. At this time the submicron aerosol particle size spectrum usually has a two–mode form (squared in Fig. 4). After the sunset and before its rise the size spectrum is single mode (crossed in Fig. 4) and the particle number density is almost constant during this period.
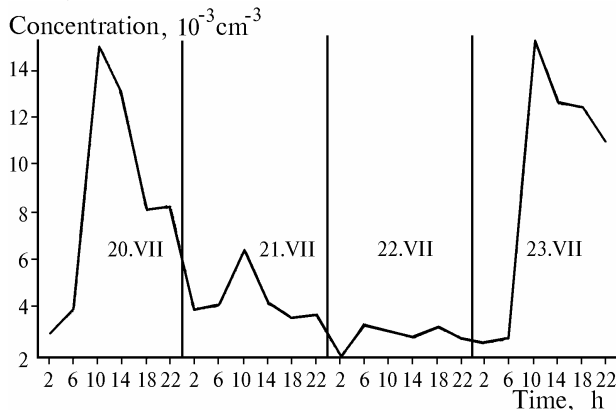
Concentration, 10$^{-3}$cm$^{-3}$

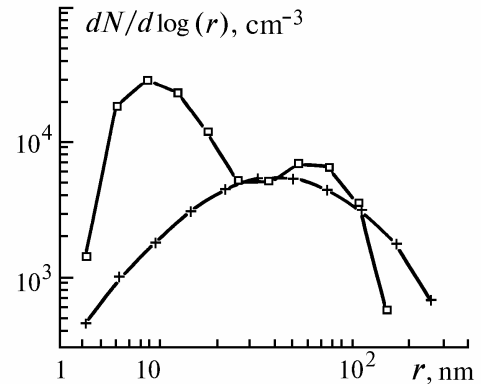*FIG. 3. Variation of aerosol particle number density as a function of time.*

*FIG. 4. Instances of aerosol particle size spectra at different time of a day.*

Let us calculate SOT using the data on the size spectrum and concentration of aerosol particles by suggesting that the particles are homogeneous in volume and spherical in form. In the estimates the size spectrum and chemical composition of aerosols are taken constant with the height increase. The vertical profile of concentration is specified by the model proposed in Ref. 10. It is represented as

$$N(z) = \begin{cases} N(0) \exp(-z/H_1), & 0 \leq z < 3.5 \text{ km}, \\ N(0) \exp(-3.5/H_1), & 3.5 \leq z < 11 \text{ km}, \\ N(0) \exp(-3.5/H_1 - z/H_2), & 11 \leq z < 30 \text{ km}, \end{cases} \quad (4)$$

$H_1 = 1.95$ km ,  $H_2 = 4.67$ km ,

where $z$ is the height of the layer in km, and $N(0)$ is the particle number density on the earth's surface.

In calculations the value $N(0)$ was assigned by formula (3) and the parameters were taken from Table II. The SOT value was calculated from the relation

$$\tau_\lambda = 0.434 \int_0^\infty \int_{r_1}^{r_2} \pi r^2 Q_{\text{ext}} (dN/dr) \, dr \, dz , \quad (5)$$

where $Q_{\text{ext}}$ is the efficiency factor of aerosol extinction which is a function of wavelength, aerosol particle size, and complex index of refraction coefficient. It was calculated by the Mie theory using the BHMIE program.[11] The atmospheric aerosols were assumed to relate to "dust–like" ones or to consist of quarts (sand soil). The complex index of refraction coefficient was taken from tables of Ref. 12 and for $SiO_2$ from Ref. 11.

The values of τ calculated by formulas (3) and (5) are listed in Table III. Both contributions to τ from different aerosol modes (different size fractions) and its total value are given here. The τ values measured experimentally at different wavelengths are listed in Table IV. The time–averaged results as well as the maximum to minimum values ratio measured during a day are given for experimental values of SOT.

It is apparent from Table III that the main contribution to SOT comes from the particles related to the second mode. The contribution from the finest (first) mode is negligible. This result is in good agreement with the studies of optical properties of atmospheric aerosols made in the other regions.[13]

*TABLE III. Calculated SOT for two types of aerosols.*

| λ, nm | Dust—like particles | | | | SiO$_2$ particles | | | |
|---|---|---|---|---|---|---|---|---|
| | Number of mode | | | Total thickness | Number of mode | | | Total thickness |
| | 1st | 2nd | 3rd | | 1st | 2nd | 3rd | |
| 383 | $2.9 \cdot 10^{-6}$ | 0.099 | $5.0 \cdot 10^{-3}$ | 0.10 | $1.5 \cdot 10^{-5}$ | 0.17 | $5.0 \cdot 10^{-3}$ | 0.17 |
| 425 | $2.9 \cdot 10^{-6}$ | 0.084 | $5.2 \cdot 10^{-3}$ | 0.089 | $1.2 \cdot 10^{-5}$ | 0.14 | $5.0 \cdot 10^{-3}$ | 0.14 |
| 491 | $<2.9 \cdot 10^{-6}$ | 0.058 | $5.1 \cdot 10^{-3}$ | 0.063 | $8.8 \cdot 10^{-6}$ | 0.11 | $5.1 \cdot 10^{-3}$ | 0.12 |
| 520 | $<2.9 \cdot 10^{-6}$ | 0.048 | $5.1 \cdot 10^{-3}$ | 0.053 | $8.8 \cdot 10^{-6}$ | 0.10 | $5.1 \cdot 10^{-3}$ | 0.11 |
| 556 | $<2.9 \cdot 10^{-6}$ | 0.039 | $5.3 \cdot 10^{-3}$ | 0.044 | $5.9 \cdot 10^{-6}$ | 0.085 | $5.1 \cdot 10^{-3}$ | 0.090 |
| 590 | $<2.9 \cdot 10^{-6}$ | 0.032 | $5.3 \cdot 10^{-3}$ | 0.038 | $5.9 \cdot 10^{-6}$ | 0.070 | $5.2 \cdot 10^{-3}$ | 0.075 |
| 672 | $<2.9 \cdot 10^{-6}$ | 0.022 | $5.2 \cdot 10^{-3}$ | 0.027 | $5.9 \cdot 10^{-6}$ | 0.046 | $5.2 \cdot 10^{-3}$ | 0.051 |
| 760 | $<2.9 \cdot 10^{-6}$ | 0.015 | $5.3 \cdot 10^{-3}$ | 0.020 | $2.9 \cdot 10^{-6}$ | 0.031 | $5.3 \cdot 10^{-3}$ | 0.037 |
| 774 | $<2.9 \cdot 10^{-6}$ | 0.014 | $5.3 \cdot 10^{-3}$ | 0.019 | $2.9 \cdot 10^{-6}$ | 0.029 | $5.3 \cdot 10^{-3}$ | 0.035 |
| 820 | $<2.9 \cdot 10^{-6}$ | 0.012 | $5.4 \cdot 10^{-3}$ | 0.017 | $2.9 \cdot 10^{-6}$ | 0.024 | $5.3 \cdot 10^{-3}$ | 0.030 |

*TABLE IV. Values of SOT measured at different wavelengths.*

| λ,nm | Date | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | July 12, 1991 | | July 13, 1991 | | July 21, 1991 | | July 22, 1991 | | July 23, 1991 | |
| | average value | max min | average value | max min | average value | max min | average value | max min | average value | max min |
| 383 | 0.060 | 1.9 | 0.145 | 1.9 | 0.264 | 1.5 | 0.098 | 1.7 | 0.060 | 1.4 |
| 425 | 0.059 | 2.8 | 0.142 | 1.9 | 0.215 | 1.3 | 0.113 | 1.6 | 0.050 | 1.2 |
| 491 | 0.055 | 2.2 | 0.120 | 2.1 | 0.215 | 1.4 | 0.094 | 1.4 | 0.042 | 1.3 |
| 520 | 0.039 | 4.0 | 0.099 | 2.3 | 0.187 | 1.5 | 0.074 | 1.5 | 0.024 | 1.6 |
| 556 | 0.042 | 2.1 | 0.098 | 2.9 | 0.194 | 1.4 | 0.080 | 1.7 | 0.040 | 1.2 |
| 590 | 0.049 | 2.3 | 0.096 | 2.9 | 0.146 | 1.4 | 0.076 | 1.8 | — | — |
| 672 | 0.046 | 2.6 | 0.097 | 2.6 | 0.174 | 1.5 | 0.074 | 1.9 | 0.044 | 1.4 |
| 760 | 0.051 | 2.4 | 0.098 | 2.2 | 0.215 | 1.1 | 0.064 | 1.9 | 0.040 | 1.2 |
| 774 | 0.038 | 3.4 | 0.074 | 4.0 | 0.201 | 1.4 | 0.054 | 2.1 | 0.028 | 1.4 |
| 820 | 0.053 | 4.4 | 0.103 | 2.1 | 0.198 | 1.4 | 0.072 | 2.0 | 0.043 | 2.0 |

The comparison of the calculated optical thickness for particles of different composition shows that variations in chemical composition are strongly manifested in optical properties of submicron fractions (the first and second models). In the spectral interval under study the optical thickness for a large—size fraction ($d > 2$ μm) of aerosols is defined by size and concentration of aerosol particles.

The theoretical analysis has shown that the SOT value in our experiments is mainly determined by characteristics of the second mode. At the same time, the calculations reveal that at constant particle number density the SOT value changes strongly as a function of size and chemical composition of aerosol particles. It is seen from the experimental data[8] that the specific feature of the second mode is an approximately constant value of particle number density. While both the size and chemical composition of these particles can change. This can be caused by condensation of either water vapors or non—volatile products formed during the gas—phase reactions occurring in the atmosphere.[14,15] When a film is formed on the surface of molecule—size particles the extinction cross—section can increase substantially.[11,14]

The selective absorption of substances forming aerosol particles can also cause nonmonotony of SOT in wavelength changing. Thus the selective absorption between 0.42 and 0.45 μm is explained by large content of hematite ($Fe_2O_3$) in aerosols.[15] Therefore in the subsequent measurements of spectral transmittance it is desirable to examine the aerosol chemical composition in more detail. To clarify the peculiarities of aerosol large—size fraction it is imperative to extend the SOT measurements to the IR spectral range.

## CONCLUSIONS

Thus based on the results of measurements of spectral optical thickness (SOT) at different wavelengths as well as concentrations and size spectrum of atmospheric aerosol carried out in July, 1991 in the southern part of lake Baikal the following conclusions can be made.

1. Variations of SOT at different wavelengths during a day are usually similar that indicates the daily constancy of the size spectrum of aerosol particles affecting SOT. The monotonous wavelength dependence of SOT was observed.

2. The aerosol particles size spectrum is well approximated by the sum of three lognormal functions. The experimental results and the theoretical analysis have shown that the optical thickness in the southern part of lake Baikal is determined by properties of the second mode of atmospheric aerosols.

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