

## INSTRUMENTS FOR MEASURING THE RADIATION ABSORPTION WITH A PHOTOMETRIC SPHERE

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*The methods and instruments available for measuring the absorption coefficients of aerosol material with the photometric sphere are reviewed. A new device in which a system of light guides is used is described in detail. An example is given in which the measurements of absorption are used to obtain data on air pollution in urban environment in different seasons.*

The instruments for measuring the most important optical parameters of aerosols – the absorption of radiation – with the use of the photometric sphere are considered in the present paper.

The absorption coefficient  $k$  can be determined with the help of independent determination of the attenuation  $a$  and scattering  $\sigma$  coefficients, which are related to each other via the formula

$$a = \sigma + \kappa.$$

Here  $a$  is determined by the well-known Bouguer's law connecting the attenuation coefficient, the radiation flux  $\Phi_0$  incident at the layer of the material with the thickness  $l$ , and the flux  $\Phi$  transmitted through this layer:

$$\Phi = \Phi_0 e^{-al} = \Phi_0 e^{-\tau},$$

where  $\tau = al$  is the optical thickness of the absorbing layer. The scattering coefficient  $\sigma$  is measured most often with the instruments of nephelometric type described in detail in Ref. 1. The coefficient  $\kappa$  can be found with an acceptable accuracy only in the case when the absorption by the medium is sufficiently high.

For improving the sensitivity and accuracy of measurements, the photometric sphere was used which had openings for the light flux and was covered from inside with the material that scattered the radiation diffusely, and had high albedo. The total area of the openings must occupy as small as possible portion of the surface area of the sphere.

One of the first papers devoted to the determination of the specific absorption with the photometric sphere (Ref. 2) was based on the measurement of the transparency  $p$  of the layer of the investigated material

$$p = \frac{\Phi}{\Phi_0} = e^{-\tau}$$

describing the light attenuation owing to scattering and absorption, and on subsequent measurement of the transparency caused solely by the absorption  $p_a$ , based on falling of the scattered light within a detector after its multiple reflection from the internal surface of the sphere.

Such way of separating the optical thicknesses was put into practice in the photometric setup, shown schematically in Fig. 1. The internal surface of the sphere consisting of two halves 1 and 2 was covered with magnesium oxide. The light flux passed through the tube 3 at the end of which the milk-matte glass 4 was inserted covered also with magnesium oxide. The hemispherical glass balloon 5 with

the investigated aerosol applied to its surface was slipped over the tube 3. The light that was multiply scattered inside the sphere fell within the FEU-79 photomultiplier 6, enclosed in the tube 7, the end of which coincided with the surface of the sphere and was shielded by the milk-matte glass 8, covered with magnesium oxide. The changeable interference filters 9 had the maximum transmission at the wavelengths  $\lambda = 405, 496, 556,$  and  $696$  nm and a bandwidth of 10–12 nm. They were placed before the photomultiplier. A FEU-62 ( $\lambda_{\text{eff}} = 977$  nm) filter and an IKS-2 filter were used for measuring in the near-IR range. At the upper part of the sphere a light source 11 emitted an additional light flux that can enter into the sphere through the tube 10.

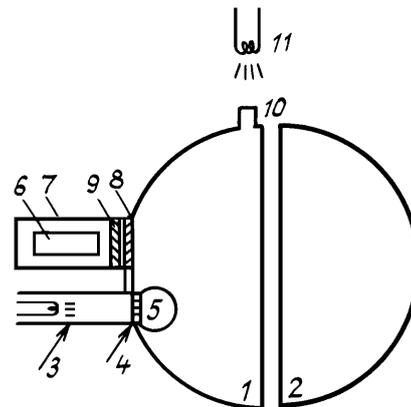


FIG. 1. The instrument for the investigation of the light absorption with the transparent balloon being sprayed with the investigated aerosol.<sup>2</sup>

Placing the balloon 5 over the tube 10, the photometric readings  $J_0$  and  $J$  were taken with and without the aerosol from which the total optical thickness of attenuation  $\tau$  was calculated. Then the balloon 5 was placed on the tube 3 with and without the aerosol and the readings  $J'_0$  and  $J'$  were obtained from which the optical thickness of absorption  $\tau_a$  was calculated. In this case redistribution of the light flux due to scattering on aerosol particles has no effect on the brightness of the walls of the sphere. The variations in the brightness could occur solely due to the absorption. Thus the readings  $J'$  and  $J'_0$  must be virtually identical if the aerosol particles did not absorb the light. This was confirmed in the experiment with the non-absorptive particles (the white powder of the ACT-4 plastics).

An account of the absorption of rays in the test sample of the aerosol due to multiple reflection from the walls of the sphere is essential in the calculation of  $\tau_a$  from measurements of  $J'$  and  $J'_0$ . The value  $\tau_a = -\ln p_a$  can be calculated if the ratio  $J'/J'_0$  is known based on the following reasons. Since the surface areas of the ends of the tubes through which light passes are small in comparison with the surface area of the sphere, in calculations we must consider only the absorption by the walls of the sphere. When the flux enter into the sphere in the absence of aerosol (balloon without dust) the brightness of the walls will be expressed as the power series  $B\rho + B\rho^2 + \dots + B\rho^n$ , where  $\rho$  is the albedo of the wall, representing geometric progression. As  $n \rightarrow \infty$ , its sum is equal to

$$J'_0 = B\rho/(1 - \rho). \quad (1)$$

Here  $B$  is the brightness of the walls of the sphere after the single reflection from the ideal sphere ( $\rho = 1$ ). The brightness of the sphere after the first act of reflection with the albedo  $\rho$  is  $B\rho$ . After the second reflection it is equal to  $B\rho^2$  after the third – to  $B\rho^3$ , and so on.

When the balloon with aerosol is placed on the light source the calculation is analogous. In this case instead of  $B\rho$  we must write  $B\rho p_a$  since the light would be attenuated after the first act of reflection at a value determined by the absorption coefficient. In the second act of reflection, when calculating the "second-order" brightness, one should take into account the fact that the light flux from the entire sphere falling within a given section of the surface partially passes through the balloon filled with aerosol. If the balloon is placed at the center so that it can be seen from any point of the sphere at the solid angle  $\Delta\omega$ , then the part of the total light flux, falling within the surface element  $\Delta S$  after absorption due to multiple scattering will be  $\Delta\omega/\pi$ , since the illumination of the surface element by the total flux is equal to  $\pi B\rho p_a$ , and by the flux transmitted through the balloon – to  $\Delta\omega B\rho p_a$ . From here, the second-order brightness is equal to

$$B\rho^2 p_a \left[ \left(1 - \frac{\Delta\omega}{\pi}\right) + \frac{\Delta\omega}{\pi} p_a^2 \right],$$

since the light passed twice through the balloon walls. Common multiplier  $p_a$  remains unchanged, since it describes the first stage – the passage of the total (not a part) incident flux through the balloon wall. Note that the first scattering act for large values of  $p_a$  plays the most important role. In the third act for the "third-order" brightness we have

$$B\rho^3 p_a \left[ \left(1 - \frac{\Delta\omega}{\pi}\right) + \frac{\Delta\omega}{\pi} p_a^2 \right]^2$$

and so on. Having found the limit of the series, we obtain

$$J' = \frac{B\rho p_a}{1 - \rho \left[ \left(1 - \frac{\Delta\omega}{\pi}\right) + \frac{\Delta\omega}{\pi} p_a^2 \right]}. \quad (2)$$

From Eqs. (1) and (2) we have

$$\frac{J'}{J'_0} = \frac{p_a(1 - \rho)}{1 - \rho \left[ \left(1 - \frac{\Delta\omega}{\pi}\right) + \frac{\Delta\omega}{\pi} p_a^2 \right]}. \quad (3)$$

The balloon had a hemispherical configuration with a diffuser at the bottom through which the main flux passed. In multiple reflection from the walls of the sphere some of rays passed through the balloon wall. This must introduce but small deviations from the results of calculations carried out on the assumption that the balloon had a spherical configuration and the rays reflected from the walls of the sphere without additional scattering by the bottom diffuser of the balloon passed through it twice. The calculations remain correct even when the balloon was placed not in the center of the sphere.

A small part of the flux in the primary act was scattered by the aerosol in backward direction and then reflected from the bottom diffuser with the balloon clamped on it was scattered again by the aerosol. A certain part of flux passed through the aerosol in the inclined directions. Therefore, the values  $\tau_a$ , determined by the proposed method are somewhat overestimated and hence give the upper limit of the influence of the aerosol absorption only. Since the light fluxes pass through the aerosol layer in various directions, we may assume that the optical thickness  $\tau_a$  determined according to formula (3) describes not vertical, but some average slant direction of the absorbing layer. Thus, the obtained values of  $\tau_a$  should be divided by a definite coefficient to reduce them to the values of the vertical directions. Starting from the dimensions of the balloon and the thickness of the aerosol layer, the average value of the optical path length  $l$  of a beam passed through the hemispherical aerosol layer can be obtained.

More perfect is the method used in Ref. 3, all the necessary calculations can then be performed in accordance with formula 3, which, as was mentioned above, is correct when the investigated aerosol is placed at the center.

The setup (Fig. 2) for measuring the attenuation and absorption coefficients of aerosol settled to the backing was designed in the form of sectional hemispheres with a diameter of 340 mm. The sphere was covered from inside with the AK-512 white deep-matte enamel with high albedo in the wavelength region 0.4 – 2.4  $\mu\text{m}$ . The FEU-51 was used as a radiation detector in the wavelength region 0.4 – 0.8  $\mu\text{m}$  and a PbS photoresistor – in the wavelength region 0.8 – 2.4  $\mu\text{m}$ . An iris diaphragm limited the incident beam. Behind the diaphragm a cassette with 12 interference light filters was set for the wavelength region 0.4 – 0.8  $\mu\text{m}$ . For measuring the optical attenuation thickness, the exit window was shielded by the diaphragm and a parallel beam transmitted through the backing 2 with the aerosol sample, was directed toward the exit window by the mirror 3, set behind the sample. When measuring the optical thickness of absorption the convergent beam passed through the backing with the aerosol and after multiple reflection gave rise to some illumination of the spherical walls which was measured with the detector 6 through the MC-20 milk-matte glass. The measurements were performed step-by-step for a clean backing and a backing with aerosol sample. In order to do this, half the backing was covered with the aerosol and the backing moved along the steel guides. When measuring the illumination of the sphere, the detector was shielded by the screen 5 in order the radiation scattered by aerosol did not fall within the detector.

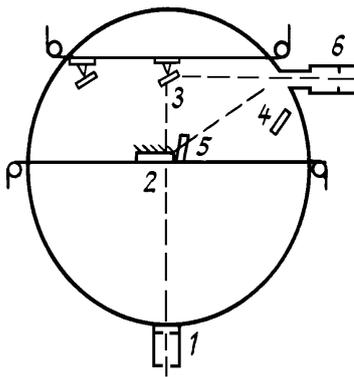


FIG. 2. The setup for the investigation of the absorption with the transparent plate placed at the center of the sphere.<sup>3</sup>

In Ref. 4 the photometric sphere was used for determining the absorption coefficient of the aerosol particles by indirect method based on the measurements of a complex part of the refractive index of the material of the investigated aerosol (Fig. 3). To this end, the continuous layer of the material was placed into the photometric sphere. The light of the xenon lamp 1 convergent with the spherical reflector 2 passed through the light filter (drawn by the black rectangle) and through the condenser 3 in the form of the parallel beam 4 was directed toward the aerosol sample 5, applied to the backing in the form of thin continuous layer. The sample was placed at the center of the photometric sphere 6. The brightness of the walls of the photometric sphere was measured by the photomultiplier 7, shielded by the screen 8 from the direct falling of light scattered on the sample. If  $\Phi$  is the light flux scattered in all directions (including the diffusely reflected flux and the flux transmitted through the sample) and  $\Phi_0$  is the flux incident on the sample from the radiation source, then the relation  $\Phi/\Phi_0$  makes it possible to determine the coefficient  $\kappa$  of the imaginary part of the complex refractive index  $m_0 = n + \kappa i$ , which is related to the absorption coefficient. Having found the values  $n$  and  $\kappa$ , the scattering and absorption coefficients of aerosol particle of a given size can be calculated according to the Mie theory. The formula for the determination of the value  $\kappa$  from the measurement of fluxes  $\Phi$  and  $\Phi_0$  has the form

$$k = -\frac{\lambda_0 S \rho^0}{4\pi m} \ln \frac{\Phi}{\Phi_0},$$

where  $\lambda_0$  is the wavelength,  $S$  is the surface area of the sample illuminated with the direct radiation,  $\rho^0$  is the density of the material of the sample and  $m$  is the mass of this material. The expression  $\frac{S\rho^0}{m}$  describes the reciprocal value of the sample thickness. All these values can be measured easily enough and, thus, the index  $\kappa$  can be determined, what was performed in Ref. 4.

When taking and preparing samples, the atmospheric aerosol undergo disturbances. The most significant changes result from vaporizing and drying the aerosol. Thus, the particles without moisture and volatile material, peculiar to the real atmospheric aerosol, are settled on the backing. Meanwhile, the specific absorption  $k/a$  and its index depend on the ratio between the volume of a given aerosol particle and its dry residue.

In this connection a set of devices based on the use of the photometric sphere, in which the investigated material occupied the entire volume of a sphere, was developed. Such devices are capable to perform the measurements of the

absorption coefficient not only for the natural aerosol but also for liquids.

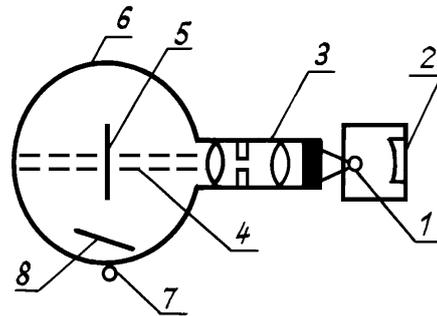


FIG. 3. The experimental setup for the investigation of the complex refractive index of solid samples.<sup>4</sup>

Expediency of using the photometric sphere as the original cavity for the absorption measurement was shown experimentally,<sup>5</sup> the advantage of the sphere consists in the fact that the beam path owing to the multiple light reflection from the walls of the sphere is lengthy enough though the diameter of the sphere is small.

The most interesting case for practice is the case when the medium has a marked (but not very significant) scattering along with the absorption. This case was discussed in Ref. 6 where it was shown that, if the absorption is insignificant, the simplified equations, given in Ref. 7, can be used for the determination of the absorption coefficient even when  $\sigma$  exceeds the absorption coefficient  $k$  by several orders of magnitude.<sup>8</sup>

When developing the correspondent experimental setups, a number of the technical problems arises. The satisfactory covering of the walls of the sphere is one of the serious problem. If the covering has high selectivity, it will lead to unacceptable large measurement errors. In this connection, the compositions with low selectivity were proposed in Ref. 9 for the surface covering of photometric spheres. In particular, the composition proposed by Azarenok et al.<sup>9</sup> on the basis of barium sulfate, polyvinyl alcohol, and ultramarine showed a high stability upon exposure of both the intense radiation and the wide range of temperatures (from 293 to 353 K).

Libin and Chernyak<sup>10</sup> used the photometric sphere to determine the scattering coefficient with the help of an original setup which they tested in studying liquids. The experimental setup consisted of the photometric sphere 5 (Fig. 4), filled with the examined medium 6. The radius of the sphere  $R$  was about 10 cm and the albedo was equal to 0.8. The light fluxes of beams 3 and 2 emitted by the single light source were alternately directed toward the cavity through the modulator 7. An exit window of the cavity 4 was placed at the center of the sphere. For  $\sigma \approx 0$ , the beam 3 was extracted from the cavity through the exit window 1 virtually without losses. The exit window has small diameter (about 1 mm) and was located in the wall of the photometric sphere. As a consequence, for  $\delta \neq 0$ , the scattered light of the beam 3 was virtually all inside the photometric sphere. The beam 2 was defocused and shifted in such a way that it did not fall within the exit window 1 and, therefore, was always inside the sphere for any value of the scattering coefficient  $\delta$ . The light that was multiply reflected from the walls of the sphere fall within the detector 9 shielded by the screen 10 in order to avoid falling of the light fluxes of the direct beams scattered in the backward direction. The dimensions of the photodetector and screen and their relative position were chosen so that the light scattered inside the sphere did not fall within the

photodetector. The ratio of the light fluxes of the beams 2 and 3 inside the cavity was equal to the ratio of readings of the photodetector, since the distribution of beams over the sphere was virtually uniform and the absorption by the material had no effect on this ratio. The photometric wedge 8, placed on the beam path 3 was used for measuring the illumination. Moving the wedge we obtained equal illuminations that could be established through vanishing the alternative component of the photocurrent.

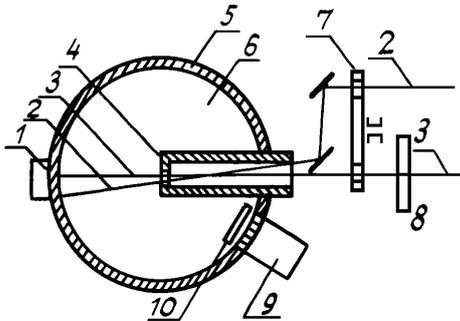


FIG. 4. Experimental setup for the investigation of the absorption with modulator and photometric wedge.

The devices in which the investigated material fill the photometric sphere have an essential drawback. When measuring the absorptance of liquids and gases, the pollution of the spherical walls, source, and other units is unavoidable that resulted in changes of the photometric properties of this device. When the albedo of the inside walls of the photometric sphere varied from 0.95 to 0.94 (i.e., slightly greater than by 1%) the measurements lead to an error in the determination of the absorption coefficient of the material under study about 40%.<sup>11</sup>

For slowly enough variations of the properties of the experimental setup, the compensation can be obtained with the help of the device described in Ref. 11. The photometric sphere, whose walls are covered with the material that diffusely reflects the light and has as large coefficient of reflection as possible, differ in the small part of its inside surface (from 1 to 3%) covered with matte-black paint having the lowest reflection coefficient. The part of this surface was shielded by the screen with high reflectance. This screen can be moved thus increasing or decreasing the area of the black surface thereby changing the effective albedo of the entire sphere. The theory makes it possible to calculate the change in the effective reflectance of the sphere  $\rho_{\text{eff}}$ , if the geometric dimensions and reflection coefficients of different parts of its surface are well known.

When the albedo changes fast, it is possible to use a device containing light guides inserted into the sphere.<sup>12</sup> The scheme of this device is drawn in Fig. 5. At the center of the photometric sphere 1 the light source 2 was placed with the balloon, having milky-matte diffusely scattered surface, whose radius was much less than the radius of the sphere  $R$ . Three light guides 3, 4, and 5 were inserted into the sphere. The input end of the light guide 3 was near the walls of the sphere and faced them, the input end of the light guide 4 was near the wall of the sphere but faced the source. The input end of the light guide 5, faced also the light source, was near the latter.

The radii of the light guides must be less than  $R$ , but much greater than the radii of the investigated particles (if, for example, the sphere was filled with aerosol or hydrosol). The input aperture of the light guide 5 must satisfy the following condition: the source area limited by the aperture

should be not more than the source area, visible from the end of the light guide. The exit ends of the light guides 3, 4, and 5 were connected to the corresponding detectors 6, 7, and 8, connected to the recording devices. The light guides were hermetically inserted in the walls of the photometric sphere.

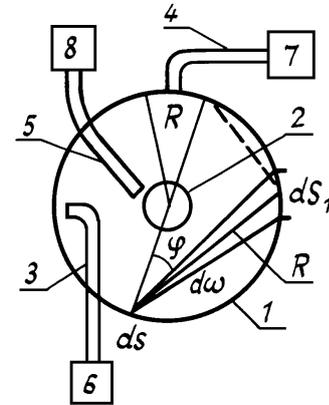


FIG. 5. Photometric sphere with the light guides.

Let us consider the operation of the device and the method of determining the absorption coefficient of the investigated media. To start with, we determine the necessary parameters of experimental setup. For this purpose the albedo of the unfilled sphere was determined first with the help of an independent method (by means of calculation of the ratio of the light flux reflected from the surface to the incident light flux measured by the same detector in the same units). Then 3 readings of the intensity received by the light guides 3, 4 and 5 were taken. Let these readings be equal  $J_1$ ,  $J_2$ , and  $J_3$ , respectively. The value  $J_1$  is proportional to the light flux  $\Phi_1$ , falling within the light guide 3 from the section of the wall of the sphere,  $J_2$  is proportional to the flux  $\Phi_2$  incident on the walls of the sphere, and  $J_3$  is proportional to the flux  $\Phi_3$  radiating by the source. Thus,  $\Phi_1 = k_1 J_1$ ;  $\Phi_2 = k_2 J_2$ ; and  $\Phi_3 = k_3 J_3$ , where the coefficients  $k_1$ ,  $k_2$ , and  $k_3$  depend on the experimental geometry, intensity of radiation, properties of the lightguides, and sensitivity of detectors. Obviously that the ratios of these coefficients  $k_1/k_2$ ;  $k_2/k_3$ ; and  $k_1/k_3$  are constant for the given experimental setup independent of the changes in the reflectance of the walls of the sphere and the bulb of the source and the transparency of the ends of the light guide (given that the ends become turbid due to sedimentation of the condensate and the degree of turbidity is identical. Obviously that fluxes  $\Phi_1$ ,  $\Phi_2$ , and  $\Phi_3$  are proportional to the illuminations  $E_1$ ,  $E_2$ , and  $E_3$  upon exposure to the corresponding fluxes. Therefore, we can write  $J_2 = C_{21} \frac{J_1}{J_1}$ , where  $C_{21}$  is constant for the given setup.

Knowing the albedo  $\rho$  and taking the readings  $J_1$  and  $J_2$ , we can find the constant  $C_{21}$ . When  $C_{21}$  is well known, we can subsequently control over the albedo of the sphere at each moment of time when situation changes and the photometric characteristics of the device change too.

Based on the value of albedo, we can find the corresponding instrumental coefficient  $C_{23}$  needed for the determination of the ratio  $E/E_0$  in the process of measurements when the situation changes. Indeed, the reading  $J_2$  is proportional to the illumination  $E$  of the walls of the

sphere upon exposure to the light flux, while  $J_3$  – to the value  $E_0 = E_3$ , i.e., to the illuminance of the walls upon exposure to the direct radiation from the source in the absence of the absorbing medium (since the end of the lightguide 5 is close to the balloon of the source, the absorption by the medium is negligible).

Thus, the calibration of the device, i.e., the determination of the coefficient  $C_{23}$  can be performed with the unfilled sphere based on the formula  $E/E_0 = C_{23}J_3/[J_2(1 - \rho)]$ , from which we have on account of the relation of photometric sphere  $E/E_0 = 1/(1 - \rho)$  that the value  $C_{23}$  can be determined from the formula  $C_{23} = J_2/J_3$ . The values which enter into the right part of the formula can be found experimentally. We hereafter use the constant  $C_{23}$  in calculating  $E/E_0$ , which in the case of absorbing medium is related to the absorption coefficient  $k$  and the radius of the sphere  $R$  by the formula

$$\frac{E}{E_0} = \frac{1}{1 - 2C_0\rho} e^{-kR}, \quad (4)$$

where  $C_0$  is given by the relation

$$C_0 = \frac{1 - (1 + 2kR)e^{-2kR}}{4R^2k^2}. \quad (5)$$

Formulas (4) and (5) are approximate. In the derivation of the formulas (see Ref. 12) it was assumed that the light from the source 2 (see Fig. 5), passing to the elementary section of the surface  $dS$  as well as coming from the elementary section  $dS$  at the angle  $\varphi$  in the solid angle  $d\omega$  to the elementary section  $dS_1$  are attenuated owing to the absorption. This assumption was also used for all successive acts of multiple reflections of beams. More exact calculational methods which take account of the actual parameters of the medium are needed for the rigorous solution of the problem. Thus, for example, it is not clear how the shape of the scattering phase function of the particles influences the final results.

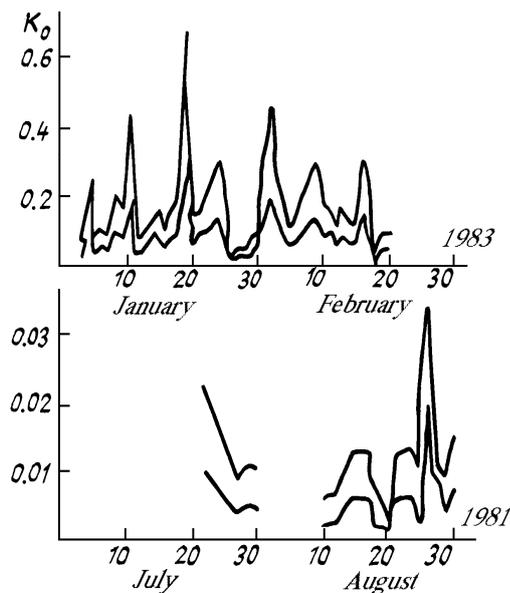


FIG. 6. Examples of the time variation in the value of  $k_s$  in winter, 1983 and summer, 1981. Upper curves correspond to  $\Lambda_s = 0.77$ , lower curves – to  $\Lambda_s = 0.88$ .

Apparently, the best method here is the Monte Carlo method successfully used to solve the problems connected with the radiation transfer in the medium with complex configuration.<sup>13</sup>

From the various applications in the field of investigation of the aerosol and hydrosol particles, the application of the above-considered instruments and methods for the control of the atmospheric pollution is of interest. As the illustration, let us give some results of investigations performed in Alma-Ata in summer 1981 and in winter 1983. The absorption coefficients of dry residue of aerosol particles  $\kappa_s$  were found. The photon survival probability  $\Lambda_s$  according to the estimates with the photometric sphere was within the limits  $0.77 < \Lambda_s < 0.88$ . In accordance with this, the upper and lower limits of the absorption coefficient of dry residue of particles were calculated at every night. The results are shown in Fig. 6. The lower curves correspond to clearer air ( $\Lambda_s = 0.88$ ) upper curves – to the most polluted air ( $\Lambda_s = 0.77$ ).

As can be seen from the figure, in winter the value of  $\kappa_s$  exceeds its summer values by an order of magnitude. The value of the absorption coefficient during the seasons can change by ten orders and more. As a consequence, identical values of  $\kappa_s$  can be observed in separate cases in winter and summer. The time variations of the value  $\kappa_s$  at night exhibit the decay of the absorption coefficient. Owing to the wind, the air by morning became several times purer. However, the air transparency at that time depends on the initial pollution and when cycles are repeated in some days with the increased of  $\kappa_s$ , the smog situations arise which can be clearly seen in the form of sharp peaks in the temporal dependence of the absorption coefficients.

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