Measurements of hydrosol scattering asymmetry by use of light field from a point source

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Received October 4, 2004

A method for measuring the asymmetry of the hydrosol scattering phase function from the ratio of irradiance generated by two sources with hemispherical direction patterns is analyzed. The method proposed was realized in ASP-15 device, installed at the place of location of the NT-200 neutrino telescope (Lake Baikal region) and measuring the optical parameters of water in the wavelength range from 350 to 690 nm. Numerical simulation of light fields by the Monte Carlo method was carried out. It is shown that, using the appropriate corrections allowing for the values of the absorption and scattering coefficients of the medium, it is possible to determine the asymmetry factor with the acceptable accuracy. A hypothesis is put forward that the real and imaginary parts of the refractive index within the chlorophyll absorption line vary in phase. This hypothesis can explain the experimentally observed decrease of the asymmetry in the range of 650–690 nm.

Introduction

Scattering phase function is an important optical parameter characterizing scattering particles that governs the process of radiation propagation through a turbid medium. For qualitative estimation of the type of medium usually it is sufficient to know the asymmetry factor K, which determines the ratio of the fluxes scattered by an elementary volume to forward and backward hemispheres:

$$K = \int_0^{\pi/2} \beta(\gamma)\sin\gamma d\gamma \Bigg/ \int_{\pi/2}^{\pi} \beta(\gamma)\sin\gamma d\gamma. \qquad (1)$$

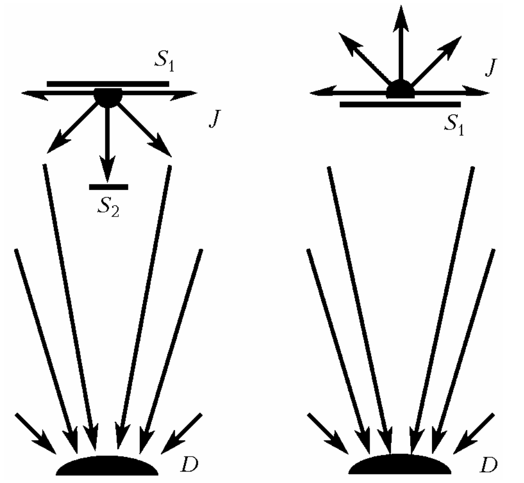
Here $\beta(\gamma)$ is the coefficient of a directed scattering, γ is the scattering angle. The values of the asymmetry factor of natural waters are determined, first of all, by the ratio of the content of large organic and fine mineral fractions in hydrosol.[1,2] According to literature data,[3,4] typical values of the asymmetry factor observed in oceans lie in the limits 8–60 for the Atlantic and 10–85 for Indian Ocean. The maximum values $K = 150$ are observed in Peru upwelling zone, which is characterized by the prevalence of large biological particles.

Precise methods for measuring the asymmetry of scattering, according to Eq. (1), should be based on illumination of the scattering volume by collimated beam and measurement of the scattered light by a narrow-angled receiver in the entire angular range from 0 to 180°. The peculiarities of long-term field experiment carried out without change of adjustment of the optical arrangement during a year require application of simple methods which do not need precise adjustment of narrow beams and do not depend on absolute calibration of the device.[5]

The methods of measuring hydrooptical parameters using radiation of an isotropically emitting source were realized in an ASP-15 stationary meter of hydrooptical characteristics (former name “Burkhan”).[6,7] It was placed near cape Ivanovskii on Lake Baikal 3.5 km far from the coast near the neutrino telescope NT-200.[8] Since 1993 the meter provides operation of the telescope by monitoring of the optical characteristics of water in the wavelength range 350 to 690 nm (absorption, scattering, scattering phase function) at the depth of 1200 m during a year. In March and April, during service maintenance of the instrumentation, measurements of hydrooptical characteristics were carried out in the entire layer from surface to bottom (1390 m).[9] The methods used to retrieve the optical parameters (absorption and scattering coefficients and scattering phase function in the angular range from 2 to 100°) are described in detail in Ref. 7.

The error in retrieving the scattering phase function in the angular range 2 to 100°, according to the estimate,[7] does not exceed 10%, that is quite acceptable for practical purposes. However, if measurements of the absorption and scattering coefficients by means of the meter of semi-spatial illumination are quite simple and were carried out in an automated mode, to measure the scattering phase function it is necessary to measure the brightness body with a narrow-angled receiver, that is more complicated in design, and does not allow one to extend measurements of the scattering phase function to the angular range above 90° because of the limited light power of the source. In this connection, a method[9] was proposed for approximate monitoring of the integral parameter, the asymmetry coefficient of the scattering phase function, using measurements of illumination from a point source with hemispherical directional pattern.

Let us consider in this paper only the principle of the method, not touching technical details of the optical layout of the device. The optical arrangement of the measurements is shown in Fig. 1. The source of isotropic radiation J is placed at the distance $R_0 \approx 5 - 10$ m from the receiver D, the meter of illumination with a cosine receiving diagram. The big screen S_1 covers the source so that only a fraction of radiation remains, directed to the side of the receiver (Fig. 1*a*) or to the back (Fig. 1*b*) hemisphere relative to the receiver. In the case shown in Fig. 1*a*, the small screen S_2 is additionally installed, which provides the shadow zone with the opening angle of 0.5° and thus removes the direct flux coming to the receiver without scattering retaining only the radiation scattered to the forward hemisphere E_F. The back-scattered flux E_B is measured according to the diagram in Fig. 1*b*.



a *b*

Fig. 1. Optical arrangement of the asymmetry factor measurements in a scattering medium.

Optical thickness of the layer between the source and the receiver $\tau = cR_0$ does not significantly exceed unity at typical values of the extinction coefficient in deep layers of water $c = 0.1–0.2$ m^{-1}, and one can expect that the contribution of multiple scattering is small. So the ratio of the radiation fluxes, initially directed forward and backward $K' = E_F/E_B$ is determined, first of all, by the asymmetry of the scattering phase function K. However, other optical parameters of the medium also affect the value K', namely the absorption a and scattering b coefficients. Can one judge about the precise value of the asymmetry factor determined by Eq. (1) from the measured value $K' = E_F/E_B$? This is just the question to be addressed in this paper.

1. Modeling of a light field in a scattering medium

The isotropic light field from a source was modeled by the Monte Carlo method using the algorithm for local estimation[10,11] for the case of non-stationary transfer equation. It was assumed that a point source $P_0(r,t) = \delta(r)\delta(t)$ of a unit intensity is placed in an infinite homogeneous scattering medium characterized by the scattering b and absorption a coefficients and the scattering phase function $g(\gamma) = \beta(\gamma)/b$. The point receiver with the cosine receiving diagram is placed at the distance of $R_0 = 5$ m from it. The received light flux is presented in the form

$$P(t) = \int_{2\pi} L(t)\cos\varphi \mathrm{d}\Omega ,$$

where $L(t)$ is the brightness at the point of receiving, and it determines the temporal shape of the signal or, in other notation, distribution of photons over the free path length $l = vt$ ($v = 0.224$ m/ns is the light speed in water). First photons reach the receiver at the moment $T_0 = R_0/v$. It is formally assumed that the receiver area is equal to 1 m^2, then the illumination at the receiving point is obtained by integrating of the flux over time:

$$E = \int_{T_0}^{\infty} P(t)\mathrm{d}t .$$

Calculations were performed for the conservative medium ($a = 0$). Then recalculation was done for the given absorption according to the known relationship[12]:

$$P(t,\ b,\ a \neq 0) = P(t,\ b,\ a = 0)\ \exp(-avt).$$

One should consider in detail the peculiarities of modeling the isotropic radiation from a source in the media with strongly anisotropic scattering. It is clear from physics of the problem that at small optical thickness of the medium (about $\tau \approx 1$) principal contribution to illumination comes from the photons initially propagating along the directions close to the direction toward the receiver. Moreover, as it will be seen below in the calculated results, the photons propagated at different angles are separated. The less is the time of a delay between the photons, the less is the range of the angles about the forward direction that form the received flux. It leads to the necessity, when modeling the initial direction of photon propagation from the source, to give preference to the photons propagating at small angles, introducing the corresponding initial weight of the photon.[13]

Two values of the scattering coefficient were taken for the calculations: $b = 0.15$ m^{-1} (characteristic of the most transparent near-surface waters of Lake Baikal in February and March) and $b = 0.015$ m^{-1} (the value, sometimes observed at the depth of 1200 m). The values of the absorption coefficient were taken in the limits 0.005–0.5 m^{-1}, that overlaps the observed range of absorption in the wavelength range 0.4–0.7 μm. Four types of the scattering phase function of sea water were selected, measured in different time by O.V. Kopelevich and V.M. Pavlov[14] (Fig. 2). Two scattering phase functions have extreme asymmetry.

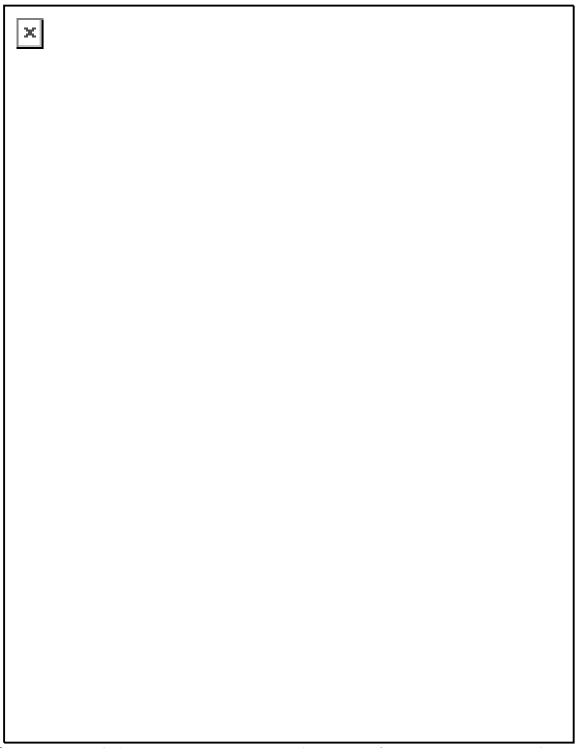


Fig. 2. Model scattering phase functions used in calculations.

The least asymmetric scattering phase function g_1 ($K = 11$, $\langle\cos\gamma\rangle = 0.788$) is observed in transparent water of Sargasso Sea, the most asymmetric g_4 ($K = 361$, $\langle\cos\gamma\rangle = 0.987$) is characteristic of the Black Sea waters. The scattering phase functions g_2 ($K = 40$, $\langle\cos\gamma\rangle = 0.924$) and g_3 ($K = 139$, $\langle\cos\gamma\rangle = 0.97$) are typically observed in the open ocean water. Besides, the molecular scattering phase function g_m was used. The scattering phase functions shown in Fig. 2 are normalized according to the condition

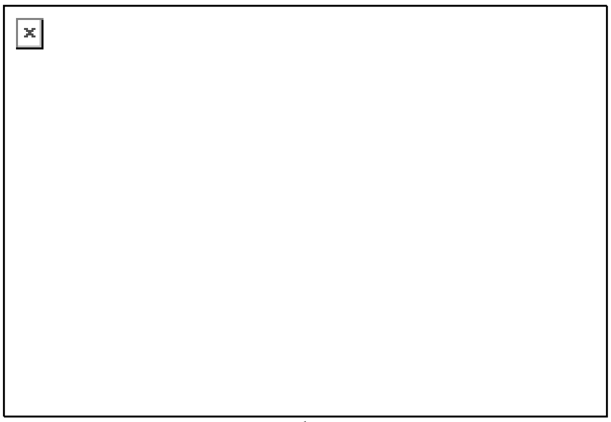
$$2\pi\int g(\gamma)\sin\gamma\,\mathrm{d}\gamma = 1.$$

The distributions of photons over the path lengths for different scattering phase functions are shown in Fig. 3*a*, and the pulse energy

$$E^t = \int_{T_0}^{t} P(t')\mathrm{d}t'$$

accumulated to the time moment t for the scattering coefficient $b = 0.15\ \mathrm{m}^{-1}$ is shown in Fig. 3*b*. The abscissa is the delay time Δt of the photons relative to the time of the first photon arrival ($T_0 = 22$ ns for $R_0 = 5$ m, hence, the time of the photons arrival $t = T_0 + \Delta t$). From 10 to 100 millions of trajectories were taken for calculations, depending on the shape of the scattering phase function, the time scale was divided into equal on logarithmic scale parts of the histogram (5 points per a decade).

a



b

Fig. 3. The shape of the photon distribution over the arrival time $P(t)$ (*a*) and the accumulated energy E^t (*b*) for the media with different scattering phase functions: g_m (*1*), g_1 (*2*), g_2 (*3*), g_4 (*4*).

The asymptotic dependence $P(t) \sim t^{-3/2}$ characteristic of an isotropic radiation[15] is shown by dotted line in Fig. 3*a* for the scattering phase functions g_m and g_4. The radiation coming to the receiver without scattering (direct beam) was not modeled, and could be calculated (if necessary) by the formula

$$E_\mathrm{dir} = \frac{1}{4\pi R_0^2}\exp(-cR_0).$$

First of all, let us pay attention to the fact that at quite long time the distribution tends to asymptotic decrease with the index $-3/2$, and statistical modeling for later moments makes no sense, because the residual energy can be calculated analytically. The moments of reaching the asymptotic t_as are marked by arrows. The more asymmetric scattering phase functions reach asymptotic later, than the less asymmetric ones. The values of the dimensionless time at the moment of reaching asymptotic $u = bvt$ are $u = 80$ for g_m and $u = 340$ for

g_4 (the value u for the first photons is equal to the optical thickness of the layer between the source and the receiver $\tau = 0.75$). The fraction of energy coming after the moment t_{as} is greater for weakly asymmetric scattering phase functions (both absolutely and relatively), it is 3.7% for g_m, 0.5% for g_1 and only 0.002% for g_4.

It is seen from Fig. 3*b* that the pulse energy is accumulated faster in media with asymmetric scattering phase function. The steps in the curves *3* and *4* of the dependence $P(t)$ at the moment $\Delta t \approx 22$ ns (delay of the photons is equal to the time of traveling through the path) are more noticeable for the asymmetric scattering phase functions g_2 and g_4 and have the shape of the second maximum, although the fraction of energy coming at this moment is insignificant. To this moment, 96% of energy comes in the medium with the scattering phase function g_4, while only 25% comes at g_m. The double-scaled temporal structure of radiation is especially noticeable for the intermediate scattering phase functions ($g_1 - g_2$). First, fast accumulation of energy occurs at the initial time moments, when scattering at small angles has been prevalent. The presence of the second maximum in the distribution $P(t)$ leads to new noticeable increase of energy, and this addition of the scattered radiation is comparable with the energy of the direct beam E_{dir}.

Undoubtedly, the delta-shaped peak of radiation at the initial time moments is caused by the prevalence of scattering at small angles at these moments, which does not lead to any noticeable increase of the photon path length. The cause of appearance of the second maximum can become clear from consideration of Fig. 4, which shows the formation of the total radiation flux by photons propagated along different directions from the source.

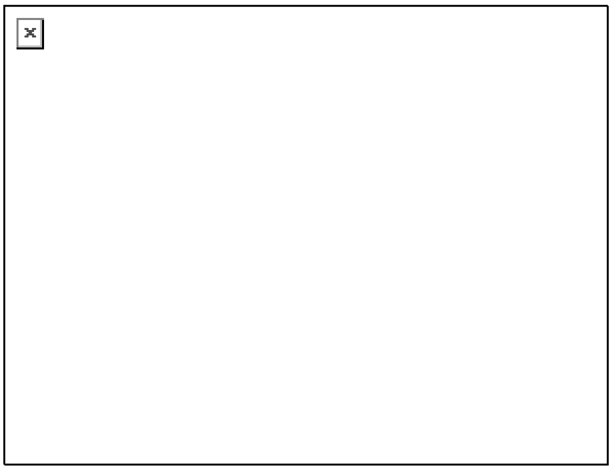


Fig. 4. The distribution $P(t)$ for the photons traveling from the source at the angles of 0° (*1*), 60° (*2*), 120° (*3*), 180° (*4*) toward the receiver.

The beams with angular divergence of 10° and different direction θ (the angle was counted from the direction from the source to the receiver) were modeled for $R_0 = 10$ m, $b = 0.15$ m^{-1} ($\tau = 1.5$) and the scattering phase function g_2. It is seen that the photons are prevalent at the initial time moments, traveling along the direction to the receiver ($\theta = 0°$, curve *1*), and at $\Delta t > 5$ ns ($\Delta u > 0.2$) along the opposite direction ($\theta = 180°$, curve *4*). All this time the contributions of the photons propagated along other directions (60°, curve *2* and 120°, curve *3*) are negligible. Only after the second maximum, when the optical length of the path has reached significant value ($\Delta t > 400$ ns, $\Delta u > 10$), the contributions of the photons propagated along different directions become equal to each other. Here 95% of energy is accumulated.

Thus, the total energy (the total flux is shown by curve *5*) is practically determined by two groups of photons, and their role is distinctly separated in time. The photons traveling forward form the peak at the initial moments, and that traveling back form the maximum at the moments when the delay has been comparable with the travel time along the path. One can conclude that the second maximum is the illumination of the back hemisphere (area behind the source). At the same time, the second maximum is even better pronounced for the radiation initially directed forward. Probably, it is formed by the photons twice scattered at the angles close to 180°. Formation of the diffuse light field, when the dependence on the direction of the photons propagation has completely disappeared and the intensity has decreased following the asymptotic law $t^{-3/2}$ (curve *6*), occurs at a small optical thickness realized in measurements $\tau \approx 1$ at a later time moments.

2. Analysis of the signals measured in the experiment

Let us consider the experimentally measured radiation fluxes, initially directed forward E_F and backward E_B.

The effect of the medium on the power distribution $P(t)$ from hemispheric sources is shown in Fig. 5 for the cases of molecular scattering (the scattering phase function g_m) and the most different hydrosol scattering phase functions g_1 and g_4. The curves for other scattering phase functions take intermediate position. Notation of the scattering phase function is indicated in the right part of the curves, dark signs show the radiation, initially directed to the forward hemisphere $P_F(t)$, and light signs show the radiation propagated to the backward hemisphere $P_B(t)$. P_F is greater than P_B at the initial time moments for all scattering phase functions. The time moment when the levels of P_F and P_B become equal is practically invariable. Then P_B begins to prevail, and the second maximum of radiation is formed. At later time moments (but before reaching the asymptotic) the fluxes become equal again, it occurs later for more asymmetric scattering phase functions. As asymmetry of the scattering phase function decreases, the difference between P_F and P_B in the region of the second maximum also decreases, and the curves for molecular scattering coincide with each other.

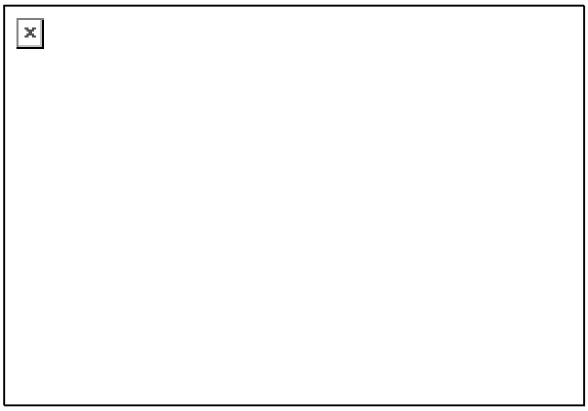


Fig. 5. The distribution $P(t)$ for the photons initially directed to the forward (dark symbols) and backward (light symbols) hemispheres. Dotted lines show the dependences taking into account absorption.

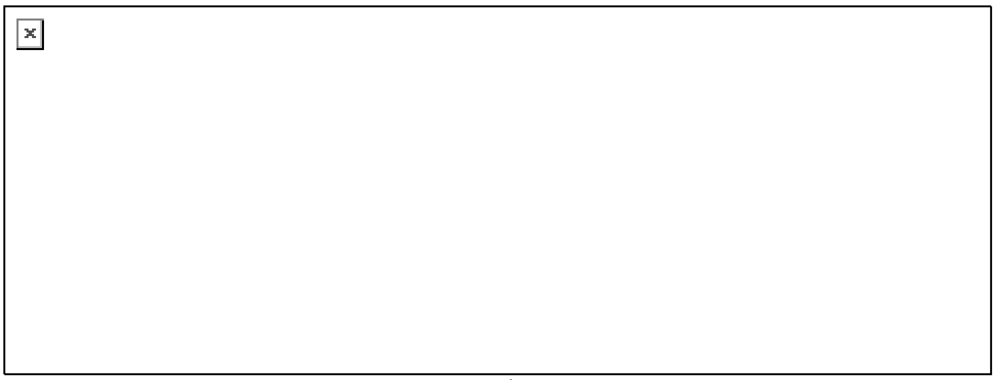
The effect of absorption on the shape of the light flux is shown in Fig. 5 by the dependence $P_F(t)$ for the scattering phase function g_4, as an example. Dotted curves correspond to the values of the absorption coefficient $a = 0.005\ \text{m}^{-1}$ (*a1*), $0.05\ \text{m}^{-1}$ (*a2*) and $0.2\ \text{m}^{-1}$ (*a3*). Gradual increase of the absorption leads, first, to weakening of the photons with long path length. Small absorption (the value $a = 0.005\ \text{m}^{-1}$ can be observed in the range 410–140 nm in clean water) leads to underestimating the most remote part of the curve $P(t)$ and weakly affects both the fluxes E_F and E_B, and their ratio. But the values $a = 0.05$–$0.1\ \text{m}^{-1}$ characteristic of natural waters strongly affect in the time interval $\Delta t = 30 - 300$ ns, to which the principal energy of the backward flux falls, while the big fraction of the flux E_F has been already accumulated. So the increase of the absorption in these limits leads to an increase of the ratio E_F/E_B. Underestimating of the received flux at the initial time moments is noticeable only at $a > 0.2\ \text{m}^{-1}$. The increase of the absorption here also leads to an increase in the measured E_F/E_B value.

Let us consider now the calculated results on the ratio of forward E_F and back E_B fluxes for the selected optical parameters. The dependence of the flux ratio $K' = E_F/E_B$ on the refractive index of the medium with the scattering phase function g_3 (the value $K = 139$) and different values of the scattering coefficient is shown in Fig. 6*a*. Curve *1* corresponds to the value $b = 0.015\ \text{m}^{-1}$, curve *2* is for $b = 0.15\ \text{m}^{-1}$. As expected, the flux ratio is not constant, but depends on other optical parameters of the medium.

Let us now try to correct the measurement results taking into account the effects connected with the influence of extinction and absorption on the parameter measured. To justify the formula (5) suggested below, the simplified diagram of formation of the measured fluxes is shown in Fig. 7*a*. The aforementioned fact is taken into account here, that two light beams determine the signal in the medium with $\tau \approx 1$.

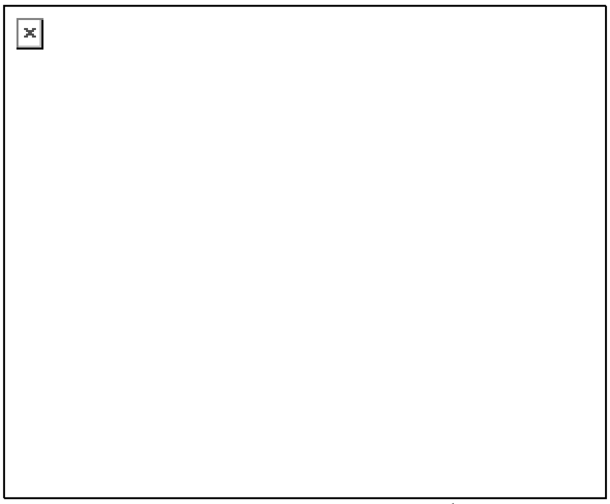
The beam, initially propagating to the side of the receiver and scattered at small angles comes at the initial time moments and forms the signal E_F. The beam directed to the opposite side from the receiver, is scattered at 180° and forms the signal E_B. In the first approximation one can neglect the contribution of photons scattered at other directions. The mean free path of the photons in the medium is determined by the extinction coefficient: $\langle l \rangle = 1/c$. Two scattering volumes (Fig. 7*a*) are at the distance $\langle l \rangle$ from the source and play the role of virtual sources of the radiation singly scattered forward (the scattering volume *FS*) and backward (the volume *BS*). Light from the back scattering volume *BS* passes an additional distance $\Delta l = 2/c$ and then causes the extinction of the flux $E_B \approx \exp(-a\Delta l)$, which should be taken into account in correcting. In the first approximation the extinction of isotropic radiation from the source is only determined by true absorption in the medium. Thus, the first stage of correction of the measured ratio $K' = E_F/E_B$ takes the form

$$K'' = K' \exp\left(-a\frac{2}{c}\right). \qquad (2)$$



a *b* *c*

Fig. 6. The dependence of the calculated flux ratio on the optical parameters of the medium (*a*) and the results of correction taking into account the extinction and absorption coefficients (*b*, *c*).



a *b*

Fig. 7. The diagram clarifying the principle of correction of the measured fluxes.

The dependence $K''(a)$ is shown in Fig. 6*b*. It is seen that curves for the media with different values of the refractive index coincide, but dependence on the absorption still remains. To take into account the effect of absorption, let us consider Fig. 7*b*, where a fragment of the dependence $P(t)$ is shown for the scattering phase function g_1 in the time interval determining the principal energy of the scattered radiation. In this range $E_B \approx \mathrm{const}(t)$, while $E_F \sim t^{-1/2}$ (dotted line). Then the flux ratio depends on the absorption:

$$\frac{E_F}{E_B} \propto \frac{\int t^{-1/2}\exp(-avt)\mathrm{d}t}{\int \exp(-avt)\mathrm{d}t} \propto \sqrt{a}. \qquad (3)$$

Thus, the correction for absorption takes the form:

$$K''' = K''/\sqrt{a}. \qquad (4)$$

The value K''' is shown in Fig. 6*c*, in which it is seen that application of the described correction in the range $a = 0.02–0.5\ \mathrm{m}^{-1}$ removes the dependence of the measured value on additional optical parameters of the medium – scattering and absorption. The final formula (in dimensionless optical coordinates) has the form:

$$K^{\mathrm{cor}} = K'\frac{\exp(2\Lambda - 1)}{\sqrt{\tau_{\mathrm{a}} + \tau/K'}}, \qquad (5)$$

where $K' = E_F/E_B$ is the measured flux ratio, $\Lambda = b/c$ is the single scattering albedo, $\tau_{\mathrm{a}} = aR_0$ is the optical thickness due to absorption. In addition to expressions (3) and (4) the term τ/K' under the root sign in the denominator of Eq. (5) removes the divergence of the result at $a \to 0$ and is essential only at very small a. The results calculated for all the scattering phase functions used, taking into account the correction (5), are shown in Fig. 8. Dotted lines correspond to the true values of asymmetry of the selected scattering phase functions, light signs are for the value $b = 0.015\ \mathrm{m}^{-1}$, dark are for $b = 0.15\ \mathrm{m}^{-1}$. It is seen that the proposed correction makes it possible to retrieve the values of asymmetry of all scattering phase functions with good accuracy, except for media with only molecular scattering, for which the accepted assumption of separation of the fluxes is not fulfilled. Deviations of the retrieved values from the true ones do not exceed 15% for the most typical scattering phase functions g_1–g_4 in the range of absorption $a = 0.02–0.5\ \mathrm{m}^{-1}$.

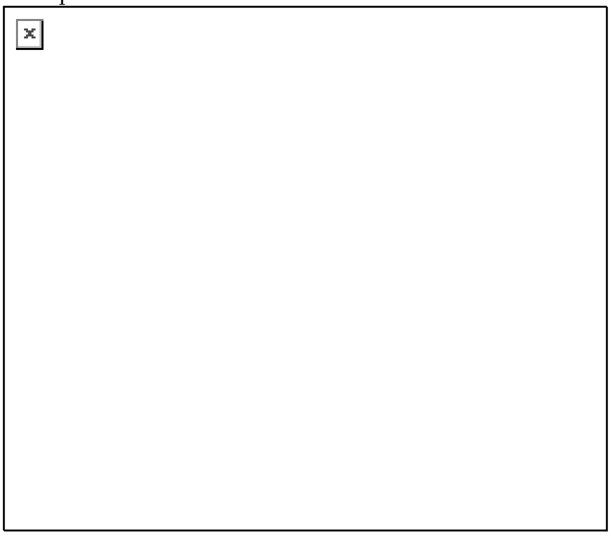


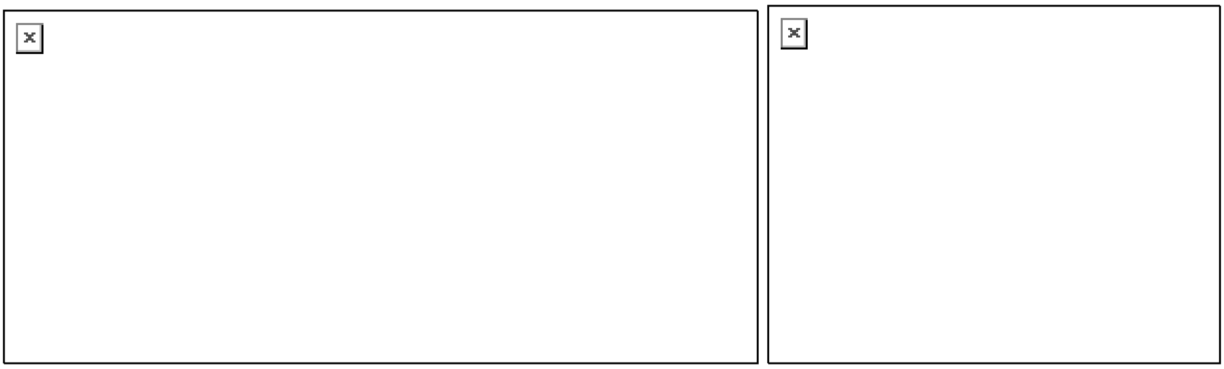
Fig. 8. The results of application of correction of the calculated values K' for the media with different scattering phase functions.

3. Discussion of the results of field observations

The data of experimental observations in the near-surface water layers of Lake Baikal carried out in March 2001 by means of an ASP-15 device are shown in Fig. 9.

The spectral dependence of absorption (Fig. 9*a*) is practically constant during the period of measurements. The values of the scattering coefficient (Fig. 9*b*) essentially increased in the period since March 21 till March 26 (curve *2* for March 26), and then again decrease. The measurement results on the asymmetry of the scattering phase functions are shown in Fig. 9*c*. Symbols show the measured values of the flux ratio $K' = E_F/E_B$, curves depict the data corrected by means of Eq. (5). The minimum in the range from 450 to 500 nm caused by minimum of absorption in this range disappears after correction, and the increase of the asymmetry of scattering is observed in the entire range from 350 to 600 nm.

Although spectral measurements of the scattering phase functions were carried out rarely, such a behavior of the asymmetry is well known[3,17] and is explained by both the decrease of the contribution of molecular scattering with the wavelength increase and by insignificant variations of the scattering phase function of large hydrosol particles.



a *b* *c*

Fig. 9. The optical parameters measured in the near-surface layers of Lake Baikal. Dates of measurements: March 17, 2001 (*1*), March 26, 2001 (*2*), March 27, 2001 (*3*).

It is seen that on March 26, together with the increase of scattering, significant increase of the asymmetry factor was observed, that can be explained by the increase in the content of large organic particles (phytoplankton) when the lake blooms. One should note that the data on the turbidity of water and the change of the scattering phase function in these days are confirmed by measurements carried out by means of the hydrosol lidar and immersed photometer – transparency meter.[18]

At the same time, no experimentally measured scattering phase functions at the wavelengths longer than 600 nm are available. The results presented here show that the dramatic decrease of the asymmetry factor to the value of the order of $K = 10$ is observed at the wavelength of 690 nm, that is usually (in observing within the range of the maximum transparency of water) characteristic of only very clean water with a low content of organic particles.

Analysis of the technical and instrumental errors of the device does not allow us to assume the existence of any specific measurement error in this wavelength range leading to underestimating the measured light fluxes by an order of magnitude. Correction of measurements by use of formula (5) can be inaccurate because of the increase of the errors in determining the optical parameters in red wavelength range. However, both corrected and measured ratios $K' = E_F/E_B$ show the decrease of the asymmetry factor in the wavelength range from 650 to 690 nm. It is especially noticeable for the media with enhanced turbidity, i.e., with the enhanced content of organic particles.

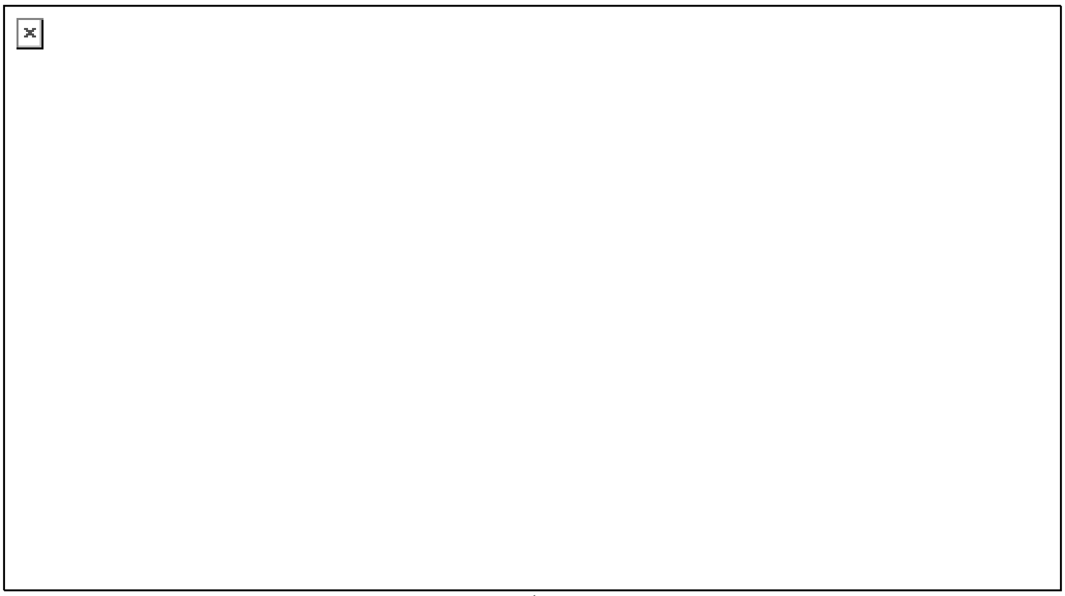
The hypothesis on the possible effect of chlorophyll absorption on the scattering phase function of hydrosol particles will be considered below in order to explain the data obtained.

4. The effect of absorption by chlorophyll on the optical parameters of hydrosol

It is known that chlorophyll A has a strong absorption line in the range of 680 nm, which can modify the spectral behavior of extinction and backscattering by sea water.[1,4] Chlorophyll is not spread homogeneously over the phytoplankton volume, but is contained in chloroplasts filling insignificant volume in the seaweed cell. For example, according to measurements,[19] the cell size of green seaweed *Chlorella vulgaris* varies in the limits between 3 and 10 μm, while the size of the inner structures (chloroplasts) has quite a narrow distribution with the mean radius of 0.8 μm with chlorophyll concentration in them of the order 0.8 kg/m^3. Imaginary part of the refractive index $m = n - i\kappa = n(1 - ia\lambda/4\pi)$ unambiguously depends on the absorption coefficient of chlorophyll A and is easily taken into account in calculations of the optical parameters of hydrosols.[19] At the same time, the dramatic variation of the real part of the refractive index is observed inside the absorption line (anomalous dispersion), which is difficult to take into account in calculations. The following value of the relative refractive index of organic particles usually assumed[1,2]: $n' = 1.02$–1.05 ($n' = n/1.34$, where 1.34 is the refractive index of water) that corresponds to the absolute value $n = 1.4$–1.47. The spectral dependence of the real (curve *1*) and imaginary (curve *2*) parts of the refractive index of chlorophyll A in the line 680 nm (Ref. 20) is shown in Fig. 10*a*.

The quick change of the real part of the refractive index from $n = 1.4$ to $n = 2.04$ at $\lambda = 700$ nm is observed simultaneously with the increase of absorption in the line, and such simultaneous change of the refractive index components should manifest itself in the scattering properties of the particle.

We have calculated the optical parameters of the chloroplasts particles, using the presented dependence of the refractive index and size distribution.[19] The calculations were carried out by accurate formulas of Mie theory.[21] An example of calculated scattering phase functions for wavelengths out of the absorption line (curve *1*, $\lambda = 600$ nm) and in the line (curve *2*, $\lambda = 670$ nm) is shown in Fig. 10*b*. The decrease of asymmetry of the scattering phase function is well seen in the absorption line at a strong increase of the backscatter.



a *b* *c*

Fig. 10. The effect of the components of the complex refractive index of chlorophyll on the scattering properties of particles in the absorption line at 680 nm.

The calculated results on the integral parameters, the scattering coefficient b (curve *1*) and the asymmetry factor K (curve *2*), are shown in Fig. 10*c*. The scattering coefficient in the line increases insignificantly. At the same time, a strong decrease of the asymmetry of the scattering phase function is observed with the change of the asymmetry factor from 65 out of the line to 10 in it. The calculated results qualitatively correspond to the experimentally observed change of asymmetry, especially that on 17th and 27th of March.

One can not consider these calculations as an accurate model of the scattering properties of phytoplankton, at least by the reason that the true chlorophyll concentration in chloroplasts is not taken into account (the 100% content of pure chlorophyll was assumed), and the real range of variations of the refractive index depends on the percentage of the chlorophyll content and on the cell size. However, the suggested mechanism of the change of the scattering phase function in the absorption line is qualitatively clear and is the following.

The values of the refractive index of chloroplast nuclei at the wavelengths out of the absorption line do not differ from that of the cell matter, and the cell scatters radiation as a soft particle (the relative value is $n' \approx 1.05$) of the size significantly greater than the wavelength. Such particles are characterized by strongly asymmetric scattering phase functions.

The real part of the refractive index of the chloroplast matter dramatically increases in the absorption line, the nuclei begin to be distinguished on the background of the cell and are independently scattering hard particle (the value of the refractive index is close to mineral particles) of the size of the order of the light wavelength. Scattering phase functions of such particles have insignificant asymmetry.

Conclusions

Simulation of operation of an ASP-15 device has shown that the method for measuring the asymmetry of scattering phase function by the ratio of illuminations from the sources with hemispheric directional pattern yields an acceptable accuracy, which allows one to monitor the variability of the scattering medium and to analyze the ratio of large and small fractions of hydrosol. The suggested correction of the measurement results requires a knowledge of the absorption and scattering coefficients of the water medium under study and is valid for all types of scattering media with the values of the asymmetry factor from 10 to 360 and the values of the absorption coefficient within the limits from 0.02–0.5 m^{-1} observed in natural waters. Simultaneous measurements of all optical parameters of water by an ASP-15 device makes it possible to refine the measured values (the absorption and scattering coefficients and the asymmetry factor) taking into account the type of the scattering medium. The unique results are obtained on the spectral behavior of the asymmetry factor of scattering in the wavelength range from 350 to 690 nm. The hypothesis is put forward about the effect of the change of the chlorophyll refractive index in the absorption line of 680 nm on the scattering properties of phytoplankton particles.

Acknowledgments

Authors would like to thank G.V. Domogatskii for the permanent attention and interest he has shown in this work.

The work was supported in part by Program No. 6.3 of the Department of Earth Sciences of Russian Academy of Sciences (project “The study of pollution of the atmosphere and near-surface waters of Baikal"), Russian Foundation for Basic Research (grant No. 04–02–16171) and the program "Universities of Russia" (No. 02.01.004).

References

1. K.S. Shifrin, *Introduction to the Ocean Optics* (Gidrometeoizdat, Leningrad, 1983), 278 pp.
2. O.V. Kopelevich, in: A.S. Monin, ed., *Ocean Optics.* Vol. 1. *Physical Optics of Ocean* (Nauka, Moscow, 1983), pp. 208–235.
3. O.V. Kopelevich, in: A.S. Monin, ed., *Ocean Optics.* Vol. 1. *Physical Optics of Ocean* (Nauka, Moscow, 1983), pp. 166–208.
4. N. Erlov, *Optical Oceanography* (Mir, Moscow, 1970), 224 pp.
5. V.I. Burenkov, B.F. Kel'balikhanov and O.V. Kopelevich, in: A.S. Monin, ed., *Ocean Optics.* Vol. 1. *Physical Optics of Ocean* (Nauka, Moscow, 1983), pp. 114–149.
6. B.A. Tarashchanskii, R.R. Mirgazov, and K.A. Pocheikin, Atmos. Oceanic Opt. **8**, No. 5, 401–403 (1995).
7. O.N. Gaponenko, R.R. Mirgazov and B.A. Tarashchanskii, Atmos. Oceanic Opt. **9**, No. 8, 677–682 (1996).
8. V. Balkanov, I. Belolaptikov, L. Bezrukov, A. Chensky, N. Budnev, I. Danilchenko, Z-A. Dzhilkibaev, G. Domogatsky, A. Doroshenko, S. Fialkovsky, O. Gaponenko, A. Garus, T. Gress, A. Karle, A. Klabukov, A. Klimov, S. Klimushin, A. Koshechkin, V. Kulepov, L. Kuzmichev, B. Lubsandorzhiev, S. Lovzov, T. Mikolajski, M. Milenin, R. Mirgazov, A. Moroz, N. Moseiko, S. Nikiforov, E. Osipova, D. Pandel, A. Panfilov, Yu. Parfenov, A. Pavlov, D. Petukhov, P. Pokhil, P. Pokolev, E. Popova, M. Rozanov, V. Rubzov, I. Sokalski, Ch. Spiering, O. Streicher, B. Tarashansky, T. Thon, R. Wischnevski, and I. Yashin, Appl. Opt. **38**, No. 33, 6818–6825 (1999).
9. N.M. Budnev, R.R. Mirgazov, and B.A. Tarashchansky, in: *Proc. of V Workshop on Phys. Processes in Natural Waters* (Limnological Inst., Irkutsk, 2000), pp. 59–62.
10. G.I. Marchuk, G.A. Mikhailov, M.A. Nazaraliev, R.A. Darbinyan, B.A. Kargin and B.S. Elepov, *Monte Carlo Method in Atmospheric Optics* (Nauka, Novosibirsk, 1976), 284 pp.
11. G.M. Krekov, G.A. Mikhailov, and B.A. Kargin. Izv. Vyssh. Uchebn. Zaved. SSSR, Fizika, No. 4, 5 (1968).
12. E.P. Zege and I.L. Katsev, *Image Transfer in a Scattering Medium* (Nauka i tekhnika, Minsk, 1985), 326 pp.
13. M.N. Gorshkov and M.A. Nazaraliev, in: G.I. Marchuk, ed., *Monte Carlo Methods in Computational Mathematics and Mathematical Physics* (CC SB AS USSR, Novosibirsk, 1979), pp. 5–14.
14. A.M. Gurfink, in: *Light Fields in Ocean* (Oceanological Institute AS USSR, Moscow, 1980), pp. 115–165.
15. E.P. Zege and I.L. Katsev, *Temporal Asymptotic Solutions of the Radiation Transfer Equation and Their Application* (Institute of Physics AS of Belarus, Minsk, 1978), 61 pp.
16. R.M. Pope and E.S. Fry, Appl. Opt. **36**, No. 33, 8710–8722 (1997).
17. V.I. Burenkov, B.F. Kel'balikhanov, and L.A. Stefantsev, in: *Abstracts of Reports. Optical Methods of the Study of Oceans and Inner Water Reservoirs*, Tallinn (1980), pp. 54–56.
18. N.M. Budnev, G.P. Kokhanenko, R.R. Mirgazov, I.E. Penner, B.A. Tarashchansky, V.S. Shamanaev, P.P. Sherstyankin, V.V. Blinov, and V.G. Ivanov, in: *Proc. of D.S. Rozhdestvensky Optical Society "Current Problems in Optics of Natural Waters,"* ed. by I. Levin and G. Gilbert, St. Petersburg (2001), pp. 318–322.
19. T. Krol, S. Kaczmarek, and O.V. Kopelevich, in: *Proc. of D.S. Rozhdestvensky Optical Society "Current Problems in Optics of Natural Waters,"* ed. by I. Levin and G. Gilbert, St. Petersburg (2001), pp. 354–359.
20. L.S. Ivlev and S.D. Andreev, *Optical Properties of Atmospheric Aerosols* (Leningrad State University, Leningrad, 1986), 360 pp.
21. D. Deirmendjian, *Electromagnetic Waves Scattering on Spherical Polydispersions* (Elsevier, New York, 1969).