Polarization characteristics of sunlight reflected and transmitted by water clouds: simple analytical solutions

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ABSTRACT

Microphysical and optical properties both warm and cold clouds are of considerable importance for climate studies. This paper is devoted to the investigation of reflection and transmission matrices of cloudy media. Simple relations between the size of droplets and the degree of polarization of reflected light are presented. The can be used for the solution of the inverse problem, namely for the determination the size of droplets from satellite polarization measurements at a global scale. The rainbow scattering region is the most promising in this respect.

1. Introduction

The influence of clouds on radiative fluxes in the terrestrial atmosphere has been studied by many authors (Kondratyev and Binenko, 1984; Minin, 1988; Liou, 1992) both experimentally and theoretically. The knowledge obtained was applied to the solution of the inverse problem, namely to the determination of the thermodynamic phase, microstructure parameters(e.g., the effective radius of droplets), single scattering albedo, optical thickness and liquid water path of cloudy media (King, 1981, 1987; Arking and Childs, 1985; Nakajima and King, 1990; King et al, 1992; Han et al, 1994; Kokhanovsky and Zege, 1996).

However, cloudy media do not only absorb and scatter radiation. They also can be considered as major sources (along with aerosols, gases and underlying surfaces) of polarized light in the terrestrial atmosphere. Clouds introduce a new property in coming solar light, namely the preferential direction of oscillation of the electric vector in a light beam. Generally speaking, both reflected and transmitted solar light becomes elliptically polarized.

The polarization arises mainly due to single scattering of a light beam by a water droplet or an ice crystal. The phenomenon of multiple scattering, which is of particular importance for clouds, leads to the randomization both photons directions and polarization states. This causes the decrease of light polarization due to increase in the optical thickness of a cloud. Another effect of multiple scattering is the introduction of ellipticity in scattered light beams. It should be pointed out that the solar light scattered by a spherical water droplet becomes partially linearly polarized. This is due to the fac

that the ellipticity of singly scattered light is equal to zero for spherical scatterers (van de Hulst, 1957).

Studies of polarization characteristics of solar light transmitted and reflected b cloudy media have a long history (Hansen, 1971a,b; Hansen and Hovenier, 1974; Coffen and Hansen, 1974; Wauben, 1992; Knibbe, 1997). However, the real burst of research in this area was given by a launch of the POLDER instrument on the board o Japanese ADEOS satellite (Deschamps et al., 1994). The POLDER was able to transmit to the Earth a huge amount of information about polarization characteristics of light reflected from cloudy media, aerosols and underlying surfaces at several wavelengths. Specifically, first three components of the Stokes vector $\vec{S}_r(I,Q,U,V)$ have been measured for wavelengths λ equal to 443, 670 and 865 nm. There is no doubt that even more advanced polarimeters with more wide spectral coverage will appear on board of different satellites in future, which makes further theoretical studies of polarization characteristics of cloudy media extremely important. This is due to potential possibilities for global retrievals of the cloud microstructure (Breon and Goloub, 1998; Kokhanovsky, 2000) , the shape of particles (Hansen, 1971a; Deschamps et al, 1994) and the optical thickness of clouds (Kokhanovsky, 2000) based on polarization measurements.

Of course, similar information can be obtained from the reflected intensit measurements. The importance of polarization lays in the possibility to check standard multispectral cloud inversion algorithms (Han et al., 1994) on the global scale. Also i could well appear that the degree of polarization can be used as a source of additiona information about the cloud particle size distributions close to the top of a cloud

(Breon and Goloub, 1998). The high proportion of photons scattered from a thin upper layer of a cloud in the creating of the light polarization is quite understandable. Multiply scattered light fluxes from deep layers are hardly polarized at all. Radiative characteristics, on the other hand, represent the cloud as a whole. Thus, the effective radius derived from radiative measurements is an average of large ensembles possibly very different particle size distributions, presented in different parts of cloudy media.

It has been emphasised by Hansen (1971a) almost 30 years ago that the fac that the polarization data refers to the cloud tops may be not a drawback but an advantage. Indeed, it is easier to interpret observations which do not refer to some average values over the entire cloud depth.

The polarization characteristics of cloudy media can be studied applying numerica codes (Hansen 1971a; de Rooij, 1985; de Haan, 1987) based on the vector radiative transfer equation solution. However, one can also use the fact that cloud fields are optically thick in most cases(King, 1987). This allows to apply asymptotic analytica relations (Domke, 1978; van de Hulst, 1980; de Rooij, 1985; Wauben, 1992), derived for optically thick disperse media with arbitrary phase functions and absorption. These solutions help us to clear physical mechanisms and main features behind the polarization change due to the increase of the size of droplets or the thickness of cloud. Analytical solutions provide also an important tool for the simplification of an inverse problem(King, 1987). They can be used, e.g., in studies of the information content of polarimetric measurements.

The drawback of well-known vector asymptotic theory for optically thick layers (Domke, 1978) is related to the fact that main equations of this theory depend on

different auxiliary functions and parameters. Calculations of these functions and parameters are rather complex. This is due to the quite general applicability of analytical asymptotic solutions in terms of the single scattering albedo.

The main task of this paper is to simplify asymptotic equations taking into account the weak absorption of light in cloudy media (King, 1981) in a broad spectra range (at least up to 2.2 μm).

We derive also the simple analytical solution for the degree of polarization of sunlight, reflected from water clouds. This relation is applied to the problem of the determination of the liquid water path and the size of droplets in cloudy media from polarimetric observations.

2. Reflection and transmission matrices.

Let us introduce the reflection $\hat{R}(\mu,\mu_0,\psi)$ and transmission $\hat{T}(\mu,\mu_0,\psi)$ matrices of cloudy media. Reflection and transmission matrices depend on the optical thickness τ of a cloud, microstructure of a cloud, cosine μ_0 of the solar angle ϑ_0, cosine μ of the viewing angle ϑ and the relative azimuth $\psi=\varphi-\varphi_0$, where φ_0 and φ are azimuths o the Sun and observer respectively. The importance of these matrices is due to the fac that they can be used for the calculation of the Stokes vector of reflected $\vec{S}_r(I,Q,U,V)$ and transmitted $\vec{S}_t(I',Q',U',V')$ light under arbitrary illumination of a cloudy layer. Namely, it follows for plane-parallel cloudy layers (Hovenier,1971):

$$\vec{S}_r(\mu,\varphi)=\frac{1}{\pi}\int_0^1 d\mu'\ \mu'\int_0^{2\pi} d\varphi'\hat{R}(\mu,\mu',\varphi-\varphi')\vec{S}_0(\mu',\varphi') \quad (1)$$

$$\vec{S}_t(\mu,\varphi)=\frac{1}{\pi}\int_0^1 d\mu' \ \mu' \int_0^{2\pi} d\varphi' \hat{T}(\mu,\mu',\varphi-\varphi')\vec{S}_0(\mu',\varphi')+\vec{S}_0(\mu,\varphi)\exp(-\frac{\tau}{\mu_0}) \quad (2)$$

where $\vec{S}_0(\mu',\varphi')$ represents the Stokes vector of the incident light beam. The second term in Eq. (2) is small for most of cloudy media $(\tau >> 1)$ and can be neglected.

In a good approximation, the Sun is a source of light incident from only one direction μ_0,φ_0. Then it follows:

$$\vec{S}_0(\mu,\varphi)=\frac{1}{\mu_0}\delta(\mu-\mu_0)\delta(\varphi-\varphi_0)\pi\vec{F}\ , \quad (3)$$

where a vector $\vec{F}$ is normalized so that the first element of $\pi\vec{F}$ is the net flux of solar beam per unit area of a cloud layer. Eqs. (1), (2) simplify if one accounts for Eq. (3):

$$\vec{S}_r(\mu,\varphi)=\hat{R}(\mu,\mu_0,\varphi-\varphi_0)\vec{F}\ , \quad (4)$$

$$\vec{S}_t(\mu,\varphi)=\hat{T}(\mu,\mu_0,\varphi-\varphi_0)\vec{F}\ , \quad (5)$$

where only first element of the column vector

$$\vec{F}=\begin{pmatrix} F_1 \\ F_2 \\ F_3 \\ F_4 \end{pmatrix} \quad (6)$$

differs from zero for unpolarized solar radiation. Thus, only elements $R_{j1},T_{j1}(j=1,2,3,4)$ are relevant to studies of solar light propagation, scattering and polarization in terrestrial atmosphere. Other elements are of interest only in the case o polarized (e.g., laser) incident beams.

One can see that the knowledge of matrices $\hat{R}(\mu,\mu_0,\varphi-\varphi_0)$ and $\hat{T}(\mu,\mu_0,\varphi-\varphi_0)$ is of primary importance for understanding radiative and polarization

characteristics of light fluxes, which emerge from a cloud body. As a matter of fact, they play a role of well-known Mueller matrices (Shurcliff, 1962; Tynes et al, 2000) for turbid media. Note, that Eq. (4) can be written in the scalar form:

$$I=R_{11}F_1, Q=R_{21}F_1,\ U=R_{31}F_1, V=R_{41}F_1 \quad (7)$$

and similarly to the transmitted light. Thus, the intensity of the reflected light is determined by the elemen R_{11} of the reflection matrix. This only element is a major concern of most cloud optics studies up to date. The degree of polarization is given b

$$P=\sqrt{P_l^2+P_c^2}\ , \quad (8)$$

where

$$P_l=\frac{\sqrt{R_{21}^2+R_{31}^2}}{R_{11}} \quad (9)$$

is the degree of linear polarization and

$$P_c=\frac{R_{41}}{R_{11}} \quad (10)$$

is the degree of circular polarization. One can show that for particular viewing geometries(e.g., $\mu_0=1, \mu=1, \varphi-\varphi_0=0,\pi$) the values of R_{31} and R_{41} are equal to zero and light is partially linearly polarized. The degree of circular polarization P_c is equal to zero in this case.

It is common to use a different definition of the degree of polarization, namely,

$$P=-\frac{Q}{I} \quad (11)$$

for linearly polarized light beams. The second component of the Stokes vector Q is proportional to the difference I_l-I_r, where values of I_l and I_r mean the intensities of light polarized parallel and perpendicular to a meridional plane, which contains the normal to a layer and viewing direction. Thus, light polarized perpendicular to the meridional plane is characterized by the positive polarization. Note, that the sign of polarization degree strongly depends on the observation geometry. For instance, it is positive in the main rainbow region(see Fig.1). It could be both positive and negative in the glory scattering region(see Fig. 1).

It should be pointed out that the Stokes vector elements I,Q,U,V (see Eq. (7)) can be used for the determination of polarization ellipse parameters such as the ellipticity and the angle of preferential oscillations of the electric vector in a light bea under study (Chandrasekhar, 1950).

Thus, reflection and transmission matrices allow to determine both intensity and polarization states of reflected and transmitted light beams under arbitrary illumination of a cloud layer. Let us study these matrices in more detail.

3. Asymptotic theory

The optical thickness of water clouds is large in most cases (Nakajima and King, 1990). Some mixed and crystalline clouds are also extended in the vertical direction for many hundred meters and could be optically thick (Uchiyama et al, 1999). Thus, the asymptotic vector theory (Domke, 1978; van de Hulst, 1980; de Rooij, 1985; Wauben, 1992) is quite applicable to cloudy media. The reflection matrix $\hat{R}(\mu,\mu_0,\varphi)$ and transmission matrix $\hat{T}(\mu,\mu_0)$ of a homogeneous plane-parallel turbid layer of a large optical thickness $\tau=\sigma_{ext}L$ (σ_{ext} is the extinction coefficient and L is the geometrical

thickness of a layer) can be written in the following form (Domke, 1978; Wauben, 1992):

$$\hat{R}(\mu,\mu_0,\psi)=\hat{R}_\infty(\mu,\mu_0,\psi)-f\hat{T}(\mu,\mu_0), \tag{17}$$

$$\hat{T}(\mu,\mu_0)=t\vec{K}(\mu)\vec{K}^T(\mu_0), \tag{18}$$

where

$$t=\frac{m\exp(-k\tau)}{1-f^2}\ . \tag{19}$$

Constant *f, m* and the vector $\vec{K}(\mu)$ can be found from following relations:

$$f=s\exp(-k\tau) \tag{20}$$

$$m=2\int_0^1 d\mu\mu\left[\vec{P}^T(\mu)\vec{P}(\mu)-\vec{P}^T(-\mu)\vec{P}(-\mu)\right], \tag{21}$$

$$\vec{K}(\mu)=m^{-1}\left[\vec{P}(\mu)-2\int_0^1 d\xi\xi\hat{R}_\infty(\mu,\xi)\vec{P}(-\xi)\right]. \tag{22}$$

Here

$$s=2\int_0^1 d\mu\mu\vec{K}^T(\mu)\vec{P}(-\mu), \tag{23}$$

and

$$\hat{R}_\infty(\mu,\xi)=\frac{1}{2\pi}\int_0^{2\pi} d\varphi\hat{R}_\infty(\mu,\xi,\psi) \tag{24}$$

is the azimuthally averaged reflection matrix $\hat{R}_\infty(\mu,\xi,\psi)$ of the semi-infinite medium with the same local optical characteristics as those for a finite layer under study. Note, that the vector $\vec{K}^T(\mu)$ is transpose to the vector $\vec{K}(\mu)$. The vector $\vec{K}(\mu)$ also occurs in the so-called Milne problem (Konovalov, 1974; van de Hulst, 1980). It describes the angular dependence and the polarization characteristics of light emerging from semi-infinite turbid layers with sources of radiation located deep inside the medium. The constant k is the smallest discrete eigenvalue, which is real and nondegenerate, obeying to the following equation:

$$(1-ku)\vec{P}(u)=\frac{\omega_0}{2}\int_{-1}^{1}d\xi\hat{Z}(u,\xi)\vec{P}(\xi), \tag{25}$$

which is called the vector characteristic equation of the radiative transfer theory. Here u is the cosine of the observation angle, ω_0 is the single scattering albedo, which is equal to the ratio of scattering and extinction coefficients, $\hat{Z}(u,\xi)$ is the azimuth-averaged phase matrix and $\vec{P}(\xi)$ is the correspondent eigenvector. The vector $\vec{P}(u)$ describes the angular distribution of light intensity and polarization characteristics o light field deep $(\tau >> 1)$ inside a semi-infinite light scattering medium. For instance, the light intensity at $\tau >> 1$ is given by $\Upsilon P_1(u)\exp(-k\tau)$, where $\Upsilon = const$.

Eqs. (17), (18) are very general and can be applied to optically thick disperse media with arbitrary local optical characteristics. However, their application to the solution of practical problems is difficult due to the necessity to solve the integra equation (25) and perform integrations (21) - (23). The reflection matrix of a semi-infinite medium $\hat{R}_\infty(\mu,\mu_0,\psi)$ obeys to the integral equation as well (de Rooij, 1985).

The problem of finding matrices $\hat{R}(\mu,\mu_0,\psi)$ and $\hat{T}(\mu,\mu_0)$ in Eqs. (17), (18) can be simplified for a broad class of so-called weakly absorbing media, which are characterized by a small value of the probability of photon absorption $\beta = 1-\omega_0$. Terrestrial clouds, e.g., fall in this category (King, 1981).

It was obtained as $\beta \to 0$ (van de Hulst, 1980; Wauben, 1992) that

$$k=\sqrt{3(1-g)(1-\omega_0)}, \tag{26}$$

$$m=\frac{8k}{3(1-g)}, \tag{27}$$

$$s=1-\frac{4k\Xi}{3(1-g)}, \tag{28}$$

and

$$\hat{R}_\infty(\mu,\mu_0,\psi)=\hat{R}^0_\infty(\mu,\mu_0,\psi)-\frac{4k}{3(1-g)}\vec{K}_0(\mu)\vec{K}_0^T(\mu_0), \tag{29}$$

where $\vec{K}_0(\mu)$ and $\hat{R}^0_\infty(\mu,\mu_0,\psi)$ are the escape vector and the reflection matrix respectively of the nonabsorbing medium with the same phase matrix as a finite layer under study,

$$g=\frac{1}{2}\int_0^\pi p(\theta)\sin\theta\cos\theta d\theta \tag{30}$$

is the asymmetry parameter of the phase function $p(\theta)$ and

$$\Xi = 3\int_0^1 d\mu\mu^2 S(\mu), \tag{31}$$

where $S(\mu)=\vec{K}_0^T(\mu)\vec{e}_1$, $\vec{e}_1$ is the unit column vector. It should be pointed out, that integral (31) is close to one for all types of phase matrices (van de Hulst, 1980; Wauben, 1992). Thus, we will neglect small differences $\Delta = 1-\Xi$ in the discussion, which follows. Note that the first moment of the function $S(\mu)$ does not depend on the phase matrix at all. This is due to the normalization condition (Wauben, 1992) :

$$2\int_0^1 d\mu\mu\ S(\mu)=1\ . \tag{32}$$

Thus, Eqs. (26) - (29) allow to reduce the complexity of Eqs. (17), (18). Onl functions $\vec{K}_0(\mu)$ and $\hat{R}_\infty^0(\mu,\mu_0,\psi)$ for a nonabsorbing semi-infinite light scattering medium should be found numerically in this more simple case. They depend on the phase matrix of a random medium in question. However, they do not depend on the single scattering albedo ω_0 and the optical thickness τ. Another interesting feature o these functions is their weak dependence on the microstructure parameters (size, shape, and chemical composition of particles) of a medium under study in the broad range of angular parameters μ,μ_0,ψ. This is due to the randomization both polarization states and propagation directions of photons in highly scattering nonabsorbing semi-infinite layers irrespectively to the local optical properties of a medium, where the propagate.

4. Approximate equations for weakly absorbing media.

Numerical calculations show that expansions (27) - (29) can be applied only for extremely narrow range of values of the probability of photon absorption $\beta = 1-\omega_0$ $(\beta < 10^{-4})$. This limits the power of the asymptotic theory for real life problems in a great extent.

Different methods to overcome this problem can be used. The most straight forward approach calls for the derivation of higher order terms in expansions (27)-(29) (Minin, 1988). However, we will use here another method, which was initially applied in the scalar radiative transfer theory (Rozenberg, 1962,1969; Zege et al, 1991; Kokhanovsky et al., 1998; Kokhanovsky, 1999). According to this alternative approach, expansions (27) - (29) are substituted by the following functions:

$$m = 1-e^{-2y}, \quad s = e^{-y}, \quad \hat{R}_\infty(\mu,\mu_0,\psi) = \hat{R}_\infty^0(\mu,\mu_0,\psi)\exp(-y\hat{W}(\mu,\mu_0,\psi)), \tag{33}$$

where

$$\hat{W}(\mu,\mu_0,\psi) = \hat{R}_\infty^{0^{-1}}(\mu_0,\mu,\psi)\vec{K}_0(\mu)\vec{K}_0^T(\mu_0), \tag{34}$$

$$y = \frac{4k}{3(1-g)} \tag{35}$$

and we assumed tha $\Xi = 1$ (see Eq. (31)).

Approximate formulae (33) transform to exact asymptotic results (27) - (29) as $y \to 0$, providing that $\Xi = 1$. However, they do allow to calculate radiative and polarization characteristics for more wide class of absorbing and light-scattering media (up to $\beta \approx 0.05 \div 0.1$, depending on the optical characteristic in question).

Thus, one can show that Eqs. (17), (18) can be presented in following forms:

$$\hat{R}(\mu,\mu_0,\psi) = \hat{R}_\infty(\mu,\mu_0,\psi) - \hat{T}(\mu,\mu_0)\exp(-y-k\tau), \tag{36}$$

$$\hat{T}(\mu,\mu_0) = t\vec{K}_0(\mu)\vec{K}_0^T(\mu_0), \tag{37}$$

where we accounted for Eqs. (33) - (35) and the approximate equality

$$m\vec{K}(\mu)\vec{K}^T(\mu_0) \approx \left(1-e^{-2y}\right)\vec{K}_0(\mu)\vec{K}_0^T(\mu_0). \tag{38}$$

The value of

$$t = \frac{1-e^{-2y}}{e^{k\tau} - e^{-2y-k\tau}} \tag{39}$$

is the global transmittance, defined as (van de Hulst, 1980)

$$t = 4\int_0^1 \mu d\mu \int_0^1 \mu_0 d\mu_0 T_{11}(\mu,\mu_0). \tag{40}$$

Eq. (40) follows from Eq. (37) accounting for the integral relation (Wauben, 1992):

$$2\int_0^1 \mu d\mu K_{01}(\mu) = 1, \qquad (41)$$

where $K_{01}(\mu)$ is the first component of the escape vector $\vec{K}_0(\mu)$.

Note, that the global transmittance, given by Eq. (39) could be transformed to the following simple form (Zege et al, 1991):

$$t = \frac{\sinh y}{\sinh(x+y)},$$

where $x = k\tau$. Eqs. (36), (37) are much simpler than the initial asymptotic formulae (17), (18). They allow to calculate the reflection and transmission matrices of thick weakly absorbing disperse media by simple means, if the solution of the problem for a semi-infinite medium with the same phase matrix as for a finite layer under consideration is available. This reduction of a general problem to the case of a semi-infinite medium is of a general importance for the radiative transfer theory.

Note, that Eqs. (36), (37) can be written in following forms:

$$\hat{R}(\mu,\mu_0,\psi) = \hat{R}_\infty^0(\mu,\mu_0,\psi)\exp[-y\hat{W}(\mu,\mu_0,\psi)]t\exp(-x-y)\vec{K}_0(\mu)\vec{K}^T{}_0(\mu_0), \qquad (42)$$

$$\hat{T}(\mu,\mu_0) = t\vec{K}_0(\mu)\vec{K}^T{}_0(\mu_0). \qquad (43)$$

Parameters x, y,and t depend on the asymmetry parameter, the single scattering albedo and the optical thickness only. They can be found from the Mie theory(van de Hulst, 1957). Matrices $\hat{R}_\infty^0(\mu,\mu_0,\psi)$, $\hat{W}(\mu,\mu_0,\psi)$ (see Eq. (34)) and the escape vector $\vec{K}_0(\mu)$ in Eqs. (42), (43) follow from the solution of the vector radiative transfer equation for semi-infinite nonabsorbing media.

It follows from Eqs. (36), (37), neglecting the polarization characteristics of a light beam under study:

$$R(\mu,\mu_0,\psi) = R_\infty(\mu,\mu_0,\psi) - T(\mu,\mu_0)\exp(-y-k\tau), \qquad (44)$$

$$T(\mu,\mu_0) = tK_0(\mu)K_0(\mu_0), \qquad (45)$$

where $T = T_{11}, R = R_{11}, R_\infty^0 = R_{\infty 11}^0, K_0 = K_{01}$ are first elements of the correspondent matrices and the vector $\vec{K}$ respectively. Eqs. (44), (45) represent well known results of the scalar radiative transfer theory (Zege et al, 1991).

Note, that formulae (42), (43) can be simplified for nonabsorbing media ($y = 0$):

$$\hat{R}(\mu,\mu_0,\psi) = \hat{R}_\infty^0(\mu,\mu_0,\psi) - \hat{T}(\mu,\mu_0), \qquad (46)$$

$$\hat{T} = t\vec{K}_0(\mu)\vec{K}_0^T(\mu_0), \qquad (47)$$

where (see Eq. (39) as $y \to 0$)

$$t = \frac{1}{1+\frac{3}{4}\tau(1-g)} \qquad (48)$$

and $\hat{R}_\infty^0(\mu,\mu_0,\psi)$ is the reflection matrix of a nonabsorbing semi-infinite medium.

It follows for the spherical albedo $r = 1-t$ of a thick cloud from Eq.(48):

$$r = \frac{\frac{3}{4}\tau(1-g)}{1+\frac{3}{4}\tau(1-g)}.$$

Note that the asymmetry parameter only slightly depends on the size of droplets in visible and $0.75(1-g) \approx 0.1$. Thus, the rough estimation of the spherical albedo of a cloud can be obtained with the following equation:

$$r = \frac{\tau}{10+\tau},$$

which gives $r = 0.8$ at $\tau = 40$.

It should be pointed out that the reflection f unction of an optically thick nonabsorbing layer can be written in the following form (see Eq. (44)) in the framework of the scalar approximation:

$$R(\mu,\mu_0,\psi)=R_\infty^0(\mu,\mu_0,\psi)-T(\mu,\mu_0), \tag{49}$$

where $T(\mu,\mu_0)=tK_0(\mu)K_0^T(\mu_0)$ and the value of *t* is given by Eq. (48). This formula can be used to retrieve the global transmittance *t*, the spherical albedo *r=1-t*, and the extrapolated optical thickness $\tau(1-g)$ (King, 1987) of cloudy media in visible. Note, that the transmission function $T(\mu,\mu_0)$ for optically thick layers does not depend on the azimuth and it follows from Eq. (40) :

$$T(\mu,\mu_0)=R_\infty^0(\mu,\mu_0,\psi)-R(\mu,\mu_0,\psi).$$

5. **The degree of polarization.**

Let us apply Eqs. (46), (47) to a particular problem, namely, to the derivation of a relation between the spherical albedo $r=1-t$ and the degree of polarization of reflected light $P(\mu_0)$ at the nadir viewing geometry (μ = 1) for a nonabsorbing light scattering medium. We will assume that a scattering layer is illuminated on the top by a wide unidirectional unpolarized light beam. The value of $P(\mu_0)$ is given simply by $-R_{12}(\mu_0,1)/R_{11}(\mu_0,1)$ (see Eqs. (7), (11)) in this case. Thus, it follows from Eq. (46), (47):

$$P(\mu_0)=\frac{P_\infty(\mu_0)}{1-U(\mu_0)(1-r)}, \tag{50}$$

where

$$U(\mu_0)=\frac{K_{01}(1)K_{01}(\mu_0)}{R_{\infty 11}(\mu_0,1)}. \tag{51}$$

The value of $P_\infty(\mu_0)=-R_{12}(\mu_0,1)/R_{11}(\mu_0,1)$ is the degree of polarization for a semi-infinite medium at the nadir observation. Note that Eq. (50) was derived with account for the equality (Wauben, 1992) : $K_{02}(1)=0$. Due to this equality, the second component of the Stokes vector for optically thick media coincides with the value of Q for semi-infinite layers at the nadir observation geometry. This fact can be used for the retrieval of the size of particles from spectral measurements o $Q(\lambda)$ even if the precise information on the optical thickness is not available. An accurate estimation of the effective radius of droplets can even be obtained from measurements o Q at a single wavelength in visible.

Both function $K_{01}(\mu_0)$ (see Fig.2) and $R^0_{\infty 11}(\mu_0,1)$ (see Fig. 3.3b in the book by Zege et al, 1991) only slightly depend on the size of scatterers. They can be represented by the following approximate formulae (Zege et al, 1991):

$$K_{01}(\mu_0)=\frac{3}{7}(1+2\mu_0), \tag{52}$$

$$R^0_{\infty 11}(\mu_0,1)=\frac{1+4\mu_0}{2(1+\mu_0)}. \tag{53}$$

Thus, it follows from Eqs. (51)-(53):

$$U(\mu_0)=\frac{54(1+2\mu_0)(1+\mu_0)}{49(1+4\mu_0)}. \tag{54}$$

The dependence $U(\mu_0)$ is presented in Fig. 3. One can see tha $U\in[1.03,1.33]$. Thus, the functi $U(\mu_0)$ does not change drastically with the angle and can be substituted by a constant value (e.g., U=1.18). Note, that the function $U(\mu_0)$ is multiplied by the global transmittance $t=1-r$ in Eq. (50), the latter being a smal number for thick clouds considered here. Thus, one can see that small errors in the function $U(\mu_0)$ do not influence very much the degree of polarization calculated with Eq. (50).

It follows from Eq. (50) that media with smaller values of the spherical albedo polarize light more effectively than media with larger values of r and $P(\mu_0) \approx P_\infty(\mu_0)/r$. The inverse proportionality between the spherical albedo r and the degree of polarization $P(\mu_0)$ was discovered experimentally by Umov(1905) almost one hundred years ago. Eq. (50) can be considered as a manifestation of this important law, which has important applications in the planetary spectroscopy (Hapke, 1993).

It follows from Eq. (50):

$$r = 1 - \frac{P(\mu_0) - P_\infty(\mu_0)}{P(\mu_0)U(\mu_0)}. \tag{55}$$

This formula can be used for the retrieval of the spherical albedo of a cloud fro polarization measurements.

The accuracy of Eq. (50) with $U=1.18$ and *r=1-t,* where t is given by Eq. (48), is studied in Fig. 4. In this Figure, data obtained from Eq. (50) and results of numerical solution of the vector radiative transfer equation for a nonabsorbing cloudy media by doubling method (van de Hulst, 1980) are presented. The value of $P_\infty(\mu_0)$ has been obtained from the exact radiative transfer calculations for a semi-infinite

layer. We have used the following particle size distribution $F(a)$ to find the phase matrix of a cloud layer:

$$F(a) = \mathrm{H} a^{\upsilon} \exp\left(-\frac{(\upsilon+3)a}{a_{ef}}\right), \tag{56}$$

where H is the normalization constant, $a_{ef} = 6\mu m$ and $\upsilon = 6$ (the cloud model Cloud C1 (Deirmendjian, 1969)). Note, that the particle size distribution (56) is normalized as follows:

$$\int_0^\infty F(a)da = 1 \tag{57}$$

The optical thickness of a cloud layer in our calculations was equal to 8, 15, and 30. Calculations of the phase matrix were performed with the Mie theory at the wavelengt 443 nm, where light absorption by water droplets can be safely neglected. The obtained value of the asymmetry parameter g (see Eqs. (30), (48)) was equal to 0.8541 in the case under investigation.

It follows from Fig. 4 that Eq. (50) indeed describes the transformation of polarization curves with the optical thickness at $\tau \ge 8$. As matter of fact this approximate equation can be applied even to media with smaller values of the optica thickness ($\tau \ge 5$, see Fig. 5). Thus, Eq. (50) can be used for the cloud optical thickness retrieval from polarimetric observations. It should be pointed out that solar angles $\vartheta_0 \ge 85^\circ$ (see Fig.4) are to be avoided because the cloud can not be modelled as a plane-parallel layer in this case.

Eq. (50) is easily generalized to account for the absorption of light in a cloud medium. Namely, it follows from Eqs. (36), (37):

$$P(\mu_0)=\frac{P^*_\infty(\mu_0)}{1-U^*(\mu_0)t\exp(-x-y)}, \qquad (58)$$

where the global transmittance t is given by Eq. (48) and

$$U^*(\mu_0)=\frac{K_{01}(1)K_{01}(\mu_0)}{R^*_{\infty 11}(\mu_0,1)} \quad . \qquad (59)$$

Values of $P^*_\infty(\mu_0)$ and $R^*_{\infty 11}(\mu_0,1)$ represent the degree of polarization and reflection function of a semi-infinite absorbing cloud at the nadir viewing geometry. Note, that Eq. (58) can be rewritten in the following form:

$$P(\mu_0)=\zeta(\mu_0,\tau)P^*_\infty(\mu_0), \qquad (60)$$

where the value o

$$\zeta(\mu_0,\tau)=\frac{1}{1-U^*(\mu_0)t\exp(-x-y)} \qquad (61)$$

can be interpreted as the enhancement factor, which is due to a finite depth of a cloud layer. It follows from Eq. (61) that $\zeta=1$ for semi-infinite layers (transmittance t is equal to zero in this case) as one can expect. It follows from Eq. (60) that zeroes o polarization curves for semi-infinite and optically thick finite layers coincide(see Eq. (60)), which is supported by numerical calculations with the radiative transfer code(see Fig.4). This is due to the fact that multiple light scattering fails to produce the polarization of incident unpolarized light. It only diminishes the polarization of singly scattered light. Thus, the angles, where the polarization is equal to zero for singly scattered light are preserved for semi-infinite layers as well.

The dependencies of the degree of polarization of a water cloud (mode *C1*) at the fixed incidence angles $\vartheta_0=30^\circ$, 37° and the observation angle $\vartheta=0^\circ$ as

functions of the probability of photon absorption $\beta=1-\omega_0$, calculated with Eq. (60) and vector radiative transfer code at $\tau=7, \lambda=443nm$, are presented in Fig.6. One can see that the accuracy of Eq. (60) is better than 2% at $\beta<0.1$ in the case under investigation. Note, that Eq. (60) a $\beta=0$ coincides with Eq. (50) as it should be.

6. Inverse problem

6.1 The sensitivity of rainbow scattering to the size of droplets

Equations obtained can be used for the parametrization of the radiative and polarization characteristics of water clouds. They can be applied to the solution of the inverse problem as well. Let us consider this last point in more detail.

It follows from Figs. 1, 7 that the degree of polarization is the most sensitive to the size of particles in the region of rainbow and glory scattering. Yet another region of high sensitivity of the degree of polarization to the size of particles is around scattering angle 90 degrees (see Fig. 1), where the polarization of light due to molecular scattering has the maximum.

The rainbow geometry is achieved if the solar angle ϑ_0, viewing angle ϑ and relative azimuth ψ satisfy to the following equation(van de Hulst, 1957; 1980):

$$-\cos\vartheta\cos\vartheta_0+\sqrt{(1-\cos\vartheta)(1-\cos\vartheta_0)}\cos\psi=\cos\theta_r,$$

where θ_r is the rainbow scattering angle, which depends on the refractive index of droplets n. The value of θ_r and is given by the following simple equation for spherical particles(van de Hulst, 1957):

$$\theta_r = \pi - 4\arcsin\sqrt{\frac{4-n^2}{3n^2}} + 2\arccos\sqrt{\frac{n^2-1}{3}}. \qquad (62)$$

One can obtain at *n=4/3:* $\theta_r \approx 138^\circ$.

It follows from Eq.(62) at $\vartheta = 0$ (nadir observation):

$$\vartheta_0 = \pi - \theta_r$$

or(see Eq. (62))

$$\vartheta_0 = 4\arcsin\sqrt{\frac{4-n^2}{3n^2}} - 2\arccos\sqrt{\frac{n^2-1}{3}}.$$

Solar rays incident at angles close to the value of ϑ_0, given by this equation will produce the rainbow phenomenon at nadir observation. Note that the angle $\vartheta_0 \approx 42^\circ$ at $n = \frac{4}{3}$.

Eq. (62) was obtained in the framework of the geometrical optics approximation, assuming tha $a/\lambda \to \infty$, where a is the radius of a droplet and λ is the wavelength(van de Hulst, 1957). It should be pointed out that the position $\theta_{\max}$ of the maximal light intensity in the region of rainbow scattering at finite ratios a/λ is related to the size of droplets and the angle θ_r by the following equation (Newton, 1966):

$$\theta_{\max} = \theta_r + \Omega(n)\rho^{-2/3},$$

where $\rho = \frac{2\pi a}{\lambda}$, $\Omega(n) = \left(\frac{3}{2}\right)^{2/3} \frac{\left(4-n^2\right)^{1/6}}{\left(n^2-1\right)^{1/2}}\Psi$ and $\Psi \approx 1.0188$. Note that $\Omega(\frac{4}{3}) \approx 2.5$.

Thus, the scattering angle for the rainbow maximum $\theta_{\max} > \theta_r$ (see Fig.1).

One can see that the the position of the maximum of the rainbow intensity and polarization depends on the rati $(a_{ef}/\lambda)^{-2/3}$(van de Hulst, 1957). Thus, it is differen for clouds with different microstructure. This dependence is confirmed also by data in Fig.7, where the degree of polarization for clouds with different size of water droplets is presented. The maximum of the degree of polarization moves to larger solar angles (at nadir observation) with the size of particles . The sensitivity of the degree of polarization to the size of droplets increases due to this fact (see Fig.7). This is o importance for the inverse problem solution.

Note, that the possibility to obtain the size of droplets from the position of secondary maxima in the region of rainbow scattering was studied in detail b Breon and Goloub(1998). It follows from Fig. 1b that the angle of the secondary maximum at $\theta \approx 155^\circ$ does not depend on the half-width parameter υ of the particle size distribution. It depends only on the effective radius of droplets(see Fig.1a). Thus, the value of a_{ef} can be retrieved from measurements of angular positions of bright rings of the rainbow pattern. However, the actual value of the degree of polarization at $\theta \approx 155^\circ$ does depend on the value of the half-width parameter υ (see Fig.1b) . This is the case for a secondary minimum at $\theta \approx 151^\circ$ (see Fig. 1b) as well. These dependencies open possibilities for the determination of the value of υ from polarimetric measurements, which is an important addition to modern retrieval algorithms, based on the reflectance spectroscopy methods(Nakajima and King, 1990).

6.2 Semi-analytical retrieval algorith

Let us study the possibility to obtain the size of particles from polarimetric measurements at the fixed viewing geometry $\vartheta_0 = 37^\circ, \vartheta = 0^\circ$), where sensitivity of measured degree of polarization to the value of the effective radius of droplets is high(see Fig.7). Results can be easily generalized for the case of other solar and observation angles, which approximately satisfy to Eq. (62).

To start with, let us rewrite Eq. (58) in the following form:

$$P(\mu_0) = \frac{P_\infty^0(\mu_0)\exp\left(yV_1(\mu_0) + y^3V_2(\mu_0)\right)}{1 - U(\mu_0)t\exp(-y\left[1 - U(\mu_0) + 0.75\tau^*\right])}, \quad (65)$$

where $\tau^* = \tau(1-g)$ is the extrapolated optical length and we accounted for approximate equalities (see Appendix 1):

$$P_\infty^*(\mu_0) = P_\infty^0(\mu_0)\exp\left(yV_1(\mu_0) + y^3V_2(\mu_0)\right), \quad (66)$$

$$R_{\infty 11}^*(\mu_0) = R_{\infty 11}^0(\mu_0, 1)\exp\left(-yU_0(\mu_0)\right), \quad (67)$$

$P_\infty^0(a_{ef}) = \Pi_1 + \Pi_2 x_{ef}^{-1/3}$

where $U = 1.18, V_1 = 1.15, V_2 = 0.06$ at $\vartheta_0 = 37^\circ$. One can find simple approximations for parameters g, y, τ in Appendix 2. The dependence of the function $P_\infty^0(37^\circ)$ on the effective size parameter at $\lambda = 443nm$ is presented in Fig. 8. It can be approximated b the following equation(see Appendix 3) :

$$P_\infty^0(a_{ef}) = \Pi_1 - \Pi_2 x_{ef}^{-1/3}, \quad (68)$$

where

$$\Pi_1 = 13.7, \quad \Pi_2 = 38.1, \quad (69)$$

$\rho_{ef} = \frac{2\pi a_{ef}}{\lambda}$ and the result is given in percent.

The real part of the refractive index of water droplets does not change considerably in the visible and near-infrared regions of the electromagnetic spectrum(Halle and Querry, 1973). Also main features of Mie scattering depend on the ratio a/λ (van de Hulst, 1957), but not separately on the size of particles and wavelength(see Appendix 3). Thus, we will assume that the dependence (68) can be also used for other wavelengths in visible and near infrared.

One can see that the degree of polarization (65) at the fixed observation geometry depends mainly on two parameters, namely on the effective size of droplets a_{ef} and the liquid water path $w = \frac{cL}{d}$, where c is the volumetric concentration of particles, d is the density of water and L is the geometrical thickness of a cloud (see Appendix 2). These two parameters $(a_{ef}$ and $w)$ can be found from spectra measurements of the degree of polarization, defined by Eq. (65).

Let us write Eq. (65) in the following form

$$P_{mes}(\lambda) = \frac{P_\infty^0(\lambda)\exp\left(y(\lambda)V_1 + y^3(\lambda)V_2\right)}{1 - Ut(\lambda)\exp(-y(\lambda)\left[1 - U + 0.75\tau^*(\lambda)\right])}, \quad (70)$$

where values of V_1, V_2, and U depend on the scattering geometry but not on the wavelength. For instance, taking two measurements of the degree of polarization a $\lambda = \lambda_1$ (in visible) and $\lambda = \lambda_2$ (in near-infrared), one obtains the system of two equations, which can form a base of the semi-analytical inversion algorithm

$$P_{mes}(\lambda_1) = \frac{P_\infty^0(\lambda_1)}{1 - Ut(\lambda_1)}, \quad (71)$$

$$P_{mes}(\lambda_2) = \frac{P_\infty^0(\lambda_2)\exp\left(y(\lambda_2)V_1 + y^3(\lambda_2)V_2\right)}{1 - Ut(\lambda_2)\exp(-y(\lambda_2)\left[1 - U + 0.75\tau^*(\lambda_2)\right])}, \quad (72)$$

where $V_1 = 1.15$, $V_2 = 0.06$ at $\vartheta_0 = 37^\circ$ and P_∞^0 is given by Eq.(68). Here we accounted for the fact that $y(\lambda_1) \approx 0$. Note, that values of $P_\infty^0(\lambda_1)$, $P_\infty^0(\lambda_2)$, and $y(\lambda_2)$ do not depend on the liquid water path by definition.

It follows from Eq. (71) that the global t ansmittance can be found from the following equation:

$$t(\lambda_1) = \frac{1}{U}\left[1 - \frac{P_\infty^0(\lambda_1)}{P_{mes}(\lambda_1)}\right]. \quad (73)$$

On the other hand, the value of $t(\lambda_1)$ is determined by Eq.(48) :

$$t(\lambda_1) = \frac{1}{1 + \frac{3}{4}\tau(\lambda_1)(1 - g(\lambda_1))}$$

or(see Appendix 2)

$$t(\lambda_1) = \frac{1}{1 + \frac{9w}{8da_{ef}}\left(1 + \frac{1.1}{\rho_{ef}^{2/3}}\right)(1 - g(\lambda_1))}.$$

Thus, it follows:

$$w = B(a_{ef})\frac{1 - t(\lambda_1)}{t(\lambda_1)}, \quad (74)$$

where (see Eq. A2.2):

$$B(a_{ef}) = \frac{8da_{ef}}{27\left(0.12 + 0.15\rho_{ef}^{-2/3} - 0.15\alpha a_{ef}\right)(1 + 1.1\rho_{ef}^{-2/3})}.$$

Here α is the absorption coefficient of water (see Appendix 2) and $t(\lambda_1)$ is determined from Eq. (73). Eq. (74) can be used for the determination of the liquid water path if the effective size of droplets a_{ef} is known. It is possible to obtain the value of a_{ef} from Eq. (72) and substitutions:

$$t(\lambda_2) = \frac{\sinh y(\lambda_2)}{\sinh(x(\lambda_2) + y(\lambda_2))},$$

$$x(\lambda_2) = \sqrt{3(1 - \omega_0(\lambda_2))(1 - g(\lambda_2))}, \; y(\lambda_2) = 4\sqrt{\frac{1 - \omega_0(\lambda_2)}{3(1 - g(\lambda_2))}},$$

$$\tau(\lambda_2) = \frac{3w}{2\rho a_{ef}}\left[1 + \frac{1.1}{(2\pi a_{ef}/\lambda_2)^{2/3}}\right], \quad \tau^*(\lambda_2) = (1 - g(\lambda_2))\tau(\lambda_2),$$

where w is determined by Eqs. (74), (73), (68) and values of ω_0 and g are defined in Appendix 2.

Thus, one can see that there is only one unknown parameter in Eq. (72) and that is the effective radius of droplets. Solving this single transcendent equation numerically, one obtains the value of a_{ef}, which subsequently can be used to find the liquid water pat w (see Eq. (74)). Knowing values of a_{ef} and w, it is easy to obtain many other parameters of cloudy media, including their transmittance, optical thickness, reflection function and spherical albedo. It should be pointed out that the degree of polarization only weakly depends on the half-width of the particle size distribution(see Fig. 1b) at the main rainbow. This simplifies the solution of the inverse problem for the effective radius retrieval.

Spec ral measurements at several wavelengths can improve the accuracy of the algorithm. However, remarkably, as we have discussed before, there is even possibility

(Kokhanovsky, 2000) to find the size of particles and liquid water path from measurements of the reflection function and the degree of polarization at the single wavelength(see Fig.7).

7. Conclusions

We presented here a system of simple equations, which can be used for parametrizations of radiative and polarization characteristics of cloudy media. The range of applicability of asymptotic theory for weakly absorbing optically thick media (see Eqs. (27) – (29)) was extended.

The algorithm of the inverse problem solution was proposed. It is based on spectral measurements of the degree of polarization of reflected light in visible and infrared at the rainbow geometry

The reflection of light from the the surface and scattering by molecules and aerosols was neglected in this study. However, they can be easily taken into account (van de Hulst, 1980; Breon and Goloub, 1998). The method described can also be used for multiple light-scattering disperse media other than water clouds. It is also applicable with slight modifications for the retrieval of the spectral dependence of the imaginary part of the refractive index of droplets, which can differ from the refractive index o pure water due to various sources of contamination.

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Appendix 1. The degree of polarization of light reflected from semi-infinite media

The task of this Appendix is to propose phenomenological analytical relations between reflection and polarization characteristics of semi-infinite plane-parallel media with weakly absorbing particles and the same characteristics for semi-infinite media with nonabsorbing particles. The phase matrix of both media is assumed to be the same. Thus, they differ only in the value of the single scattering albedo $\omega_0 = 1-\beta$.

It follows from Eq. (33) in the scalar case:

$$R_\infty(\mu,\mu_0,\psi) = R_\infty^0(\mu,\mu_0,\psi)\exp(-yU(\mu,\mu_0,\psi)), \qquad \text{(A1.1)}$$

where

$$U(\mu,\mu_0,\varphi) = R_\infty^{0^{-1}}(\mu_0,\mu,\varphi)K_0(\mu)K_0(\mu_0) \quad . \qquad \text{(A1.2)}$$

The dependence (A1.1) at $\vartheta_0 = 0^\circ, \vartheta = 37^\circ, U = 1.18$ is presented in Fig. A1.1 together with results obtained with exact calculations. One can see that the accuracy of Eq. (A1.1) is high up to $\beta = 0.05$. Note, that Eq. (29) can be applied only a $\beta < 0.001$.

The same type of approximation can be used for the degree of polarization:

$$P_\infty(\mu,\mu_0,\psi) = P_\infty^0(\mu,\mu_0,\psi)\exp(yV_1(\mu,\mu_0,\psi)), \qquad \text{(A1.3)}$$

where $V_1 = 1.15$ at $\vartheta_0 = 0^\circ, \vartheta = 37^\circ$. However, this equation has a poor accuracy a $\beta \geq 0.02$ (see Fig.A1.2), where the following modified formula should be applied:

$$P_\infty(\mu,\mu_0,\psi) = P_\infty^0(\mu,\mu_0,\psi)\exp\left(yV_1(\mu,\mu_0,\psi) + y^3V_2(\mu,\mu_0,\psi)\right), \quad \text{(A1.4)}$$

where $V_1 = 1.15, V_2 = 0.06$ at $\vartheta = 0^\circ, \vartheta_0 = 37^\circ$. Note, that constants V_1, V_2 are found by fitting Eq. (A1.4) with results of the vector radiative transfer equation solution. The accuracy of Eq.(A1.4) is studied in Fig. A1.3. It is high up to $\beta = 0.1$. It should be pointed out that Eq.(A1.1) can be also modified in a full analogy with Eq. (A1.4) to improve its accuracy at $\beta > 0.05$.

Appendix 2. Local optical characteristics of cloudy media

The extinction coefficien σ_{ext}, the asymmetry parameter g, and the absorption coefficient σ_{abs} of cloudy media can be found from the following geometrical optics equations, which account also for wave corrections (Kokhanovsky, 1999):

$$\sigma_{ex} = \frac{3c}{2da_{ef}}\left(1 + \frac{1.1}{(\rho_{ef})^{2/3}}\right) \quad \text{(A2.1)}$$

$$g = 0.88 - \frac{1}{2(\rho_{ef})^{2/3}} + 0.15\alpha a_{ef}, \quad \text{(A2.2)},$$

$$\sigma_{abs} = 1.25\alpha\left(1 - \alpha a_{ef}\right)\left(1 + 0.34(1 - \exp(-8\lambda / a_{ef}))\right), \quad \text{(A2.3)}$$

where the effective radius a_{ef} is equal to the ratio of third and second moments of the particle size distribution, $\rho_{ef} = 2\pi a_{ef} / \lambda, \alpha = 4\pi\chi / \lambda$, c is the liquid water content, d is the density of water, χ is the imaginary part of the refractive index of particles and λ is the wavelength. It was assumed that the real part of the refractive index of water is equal to *1.34.* Thus, the spectral dependence $n(\lambda)$ was neglected. Note, that Eqs. (A2.1) - (A2.3) can be used to calculate the single scattering albedo

$$\omega_0 = 1 - \frac{\sigma_{abs}}{\sigma_{ext}} \quad \text{(A2.4)}$$

and the similarity parameter

$$y = 4\sqrt{\frac{1-\omega_0}{3(1-g)}}. \quad \text{(A2.5)}$$

The simple expression for the optical thickness $\tau = \sigma_{ext}L$ follows from Eq. (A2.1):

$$\tau = \frac{3w}{2da_{ef}}\left(1 + \frac{1.1}{(\rho_{ef})^{2/3}}\right),$$

where w=cL is the liquid water path.

Comparisons with the Mie theory show that the accuracy of Eq. (A2.1) is better than 5% for water droplets with the effective radius in the range 4-16 micrometers at λ < 2.2 μm. It is up to 10% in the same spectral range for values of the absorption coefficient and co-asymmetry parameter *1-g*. It is smaller than 5% for both *1-g* and σ_{abs} at λ < 1.2 μm. The dependence of $y(\lambda)$ obtained with Eqs. (A2.1)-(A2.5) and Mie calculations for spherical polydispersions of water droplets with the gamma particle size distribution (57) is presented in Fig. A2.1 at effective radii $a_{ef} = 4\mu m$ and $a_{ef} = 16\mu m$. The half-width parameter of the particle size distribution υ was equal to 6. The data for $\chi(\lambda)$ were taken from Halle and Querry(1973). One can see that the accuracy of Eqs. (A2.1)-(A2.3) is high. The error is smaller than 7% up to $\lambda = 1.8\mu m$. The error o geometrical optics equations (A2.2) and (A2.3) increases for clouds with small $(a_{ef} < 4\mu m)$ and large $(a_{ef} > 16\mu m)$ droplets. This is due to limitations $\rho_{ef} >> 1$ and $\alpha\rho_{ef} << 1$, used for derivations of Eqs. (A2.2) and (A2.3) (Kokhanovsky, 1999). However, very small and large values of average radii of droplets are of a rear occurrence in natural water clouds (Han et al, 1994).

Appendix 3. Degree of polarization of singly scattered light at the rainbow maximum

The degree of polarization of light singly scattered by a droplet with the radius a can be found from the following general equation:

$$P = \frac{G_1 - G_2}{G_1 + G_2}, \tag{A3.1}$$

where $G_j = \frac{4i_j}{\rho^2}$ (*j=1,2*) is the gain relatively to an isotropic scattering (van de Hulst, 1957), $\rho = \frac{2\pi}{\lambda}a$, i_j is the dimensionless Mie intensity (van de Hulst, 1957). Note, that the index *j* characterizes the state of polarization.

The size of droplets in clouds is much larger than the wavelength λ of the incident solar light. Thus, the ray optics can be used for the calculation of intensities i_j and gains G_j instead of the Mie theory. The ray optics, however, breaks down for light rays singly internally reflected inside in droplet and propagated after the scattering event in the direction specified by angles close to the value of the rainbow angle (see Eq. (62)). The function $G_j(x,\theta)$ can be presented in the following general form at the main rainbow (van de Hulst, 1957):

$$G_j(\rho,\theta) = G_j^*(\rho,\theta) + G_j^R(\rho,\theta), \tag{A3.2}$$

where $G_j^R(\rho,\theta)$ is the gain due to rainbow rays and $G_j^*(\rho,\theta)$ represents the contribution of purely geometrical optics scattering (e.g., the reflection of light by the surface of a droplet) in the scattering direction specified by the angle θ. This contribution does not depend on the size of droplets in the spectral regions, where water can be considered as a nonabsorbing medium.

One can obtain at the angle of the rainbow maximum $\theta_{\max}$ (van de Hulst, 1957):

$$G_j^R(x,\theta_{\max}) = \Phi_j(n)\rho^{1/3}, \tag{A3.3}$$

where $\Phi_j(n)$ is the well-known function (van de Hulst, 1957) of the refractive index of particles *n*. Thus, it follows from Eqs. (A3.1)-(A3.3):

$$P(\theta_{\max}) = \frac{A + B\rho^{1/3}}{M + N\rho^{1/3}}, \tag{A3.4}$$

where $A = G_1^* - G_2^*$, $B = \Phi_1 - \Phi_2, M = G_1^* + G_2^*$, $\mathrm{N} = \Phi_1 + \Phi_2$. Eq. (A3.4) can be used for parametrizations of the degree of polarization of singly scattered light at the maximum of rainbow. This equation is simplified taking into account tha $\frac{M}{N\rho^{1/3}} << 1$ at large size parameters:

$$P(\theta_{\max}) = \Theta_1 - \Theta_2 x^{-1/3}, \tag{A3.5}$$

where $\Theta_1 = A/N, \Theta_2 = -B/N$. One can see that the degree of polarization at the main maximum of the rainbow can be approximated by a linear function of the parameter $\Omega = \rho^{-1/3}$ (or $\Omega_{ef} = \rho_{ef}^{-1/3}$ for spherical polydispersions).

It is safe assume that the general dependence (A3.5) is valid for semi-infinite nonabsorbing media as well. This is due to the fact that the polarization of light in cloudy media is due to single scattering of photons by droplets. The multiple light scattering reduces the degree of polarization practically not changing its general dependence on the size of droplets. It means that values of Θ_1 and Θ_2 will be smaller than in the case of single light scattering. Numerical calculations (see Fig. 8) show tha

Eq. (A3.5) do can be applied for the calculation of the degree of polarization in the vicinity of the maximum of the main rainbow for semi-infinite nonabsorbing meida.

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FIGURE CAPTIONS

Fig.1a The degree of polarization of light scattered by a polydispersion of spherical water droplets characterized by the gamma particle size distribution described by Eq. (56) with the half-width parameter $\upsilon = 6$ and the effective radius $a_{ef} = 4,6,8,10,$ and 16 micrometers as the function of the scattering angle at $\lambda = 443nm$.

Fig.1b The same as in Fig. 1a but at $a_{ef} = 6$ micrometers and different values of the half-width parameter $\upsilon = 2,4,6,8,10,12$.

Fig. 2 The dependence of the escape function K_{01} on the cosine of the observation angle for the Henyey-Greenstein phase function (Yanovitskij, 1997) with the asymmetry parameter $g = 0,0.5,0.75,0.8$. Results of calculations with Eq. (52) are presented by a dashed line.

Fig.3 The dependence $U(\vartheta_0)$ according to Eq.(53). The dashed line represents the average value of $U = 1.18$.

Fig.4 The degree of polarization of light reflected from a water cloud at nadir observation (the cloud model C1) at $\lambda = 443nm$ and different values of the optical thickness $\tau = 8,15,30$ according to exact calculations (lines) and Eq. (50) at $U = 1.18$. (symbols).

Fig.5 The degree of polarization of a water cloud (model C1) at $\lambda = 443nm$, $\vartheta = 0^\circ$, $\vartheta_0 = 37^\circ$ as the function of the inverse optical thickness, calculated with Eq. (50) at $U = 1.18$ and exact calculations with a doubling code(symbols).

Fig.6 The degree of polarization of a water cloud (model C1) at $\lambda = 443nm$, $\vartheta = 0^\circ, \vartheta_0 = 30^\circ, 37^\circ, \tau = 7$ as a function of the probability of photon absorption, calculated with the exact radiative transfer code(lines) and Eq.(60)(symbols).

Fig.7 The degree of polarization of light reflected from a semi-infinite water cloud at nadir observation and $\lambda = 443nm$ and different values of the effective radius $a_{ef} = 4, 6, 8, 12, 16\mu m$, obtained with the exact code. The half-width parameter of the particle size distribution is equal to 6 .

Fig. 8. The dependence of the degree of polarization of semi-infinite cloud on the effective radius of particles a $\lambda = 443nm$, $\vartheta = 0^\circ, \vartheta_0 = 37^\circ$ according to exact calculations (symbols) and Eq. (68).

Fig. A1.1 The dependence of the reflection function of the semi-infinite cloudy media (model Cloud C1) according to exact calculations (circles), Eq. (A1.1)(approximate, new) and Eq. (29) (approximate, old).

Fig. A1.2 The same as in Fig.A1.1 but for the degree of polarization(see Eq.(A1.3)(approximate, new) and Eq. (29) (approximate, old).

Fig. A1.3 The same as in Fig. A1.2, but the approximate Eq. (A1.4) has been used.

Fig. A2.1 The dependence of y on the wavelength according to exact Mie calculations for a polydispersions of spherical water droplets with the same particle size distribution as in Fig.1. The data obtained with Eqs. (A2.1)-(A2.5) are also presented.