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Spectral polarimetric light-scattering by particulate media: 1. Theory of spectral Vector Radiative Transfer

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ABSTRACT

Spectral polarimetric light-scattering by particulate media has recently attracted growing interests for various applications due to the production of directional broadband light sources. Here the spectral polarimetric light-scattering signatures of particulate media are simulated using a numerical model based on the spectral Vector Radiative Transfer Equation (VRTE). A microphysical analysis is conducted to understand the dependence of the light-scattering signatures upon the microphysical parameters of particles. We reveal that depolarization from multiple scattering results in remarkable spectral and directional features, which are simulated by our model over a wide spectral range from visible to near-infrared. We propose to use these features to improve the inversion of the scattering problem in the fields of remote sensing, astrophysics, material science, or biomedical.

1. Introduction

Multiple light-scattering refers to electromagnetic scattering by collections of particles from ultraviolet to infrared and occurs in particulate media when light is scattered several times before detection [1–3]. It is prominent when light propagates through dense media such as plumes, clouds, or colloids. This phenomenon has been an object of interest in many scientific fields from remote sensing [4], astrophysics [5] to biomedical [6] and material science [7].

Several inverse methods have been developed to retrieve microphysical properties of particulate media [8]. However, the inversion of single-wavelength scattering signatures is usually not permitted without prior assumptions. Such restrictions are due to the non-uniqueness in the solutions of the scattering problem [9]. Prior knowledge about particles (*e.g.* refractive index) is usually necessary to retrieve microphysical properties. Polarimetric [10] or multispectral measurements [11–13] have been proposed as a useful tool to improve light-scattering inverse methods since they provide additional information.

Recent experiments have reported the potential use of merging spectral and polarimetric light-scattering for optical diagnostic [14–19]. Due to the ill-posed inverse scattering problem, we propose to merge spectral (*i.e.* broadband or hyperspectral) and polarization information in order to develop novel inversion methods. A numerical model based on the spectral Vector Radiative Transfer Equation (VRTE) is presented to compute the spectral polarimetric light-scattering by particulate media. Furthermore, a microphysical analysis based on our model is conducted to determine the role and dependence of the particles properties (*i.e.* mean size, refractive index, and volume fraction) on simulated signatures. This approach is expected to improve physical insights and remove solution ambiguities in the development of future inverse methods.

2. Theory

The radiative transfer theory is applied with great results in astrophysics, remote sensing, physics, geophysics, atmospheric physics, and other areas of science. The Radiative Transfer Equation (RTE) was first introduced

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in 1905 for atmospheric studies [20] and solved in 1943 using the invariance principle for the problem of diffuse reflection of light by a scattering medium [21]. In this theory, particulate media are modeled as semi-infinite plane–parallel multi-layered isotropic scattering media to account multiple-scattering. One essential condition for using the RTE to model multiple light-scattering by particulate media is that the distance d between particles must be larger than the wavelength λ such as $d > \lambda$. In such a way, the scattering events always occur in the far-field region of each particle and prevent coherent effects such as dependent scattering.

Later, this theory has been extended to a larger range of scientific problems, including anisotropic scattering medium and electromagnetic scattering by an ensemble of randomly distributed and oriented particles [22]. The Vector Radiative Transfer Equation (VRTE) [23] is the vector counterpart of the RTE and has been recently derived from first-principles [24].

Let us first consider a particulate media as an ensemble of N particles in random orientations and positions in a non-scattering host medium. The spectral VRTE is given for a polarized broadband directional light source as:

$$\mathbf{u}\cdot\nabla\mathbf{S}(\mathbf{r},\mathbf{u},\lambda) = -\mathbf{k}_{ext}(\mathbf{r},\lambda)\cdot\mathbf{S}(\mathbf{r},\mathbf{u},\lambda)$$
$$+\frac{1}{4\pi}\int_0^{2\pi}\int_0^1 d\mathbf{u}'\cdot P(\mathbf{r},\lambda)\cdot\mathbf{k}_{sca}(\mathbf{r},\mathbf{u},\mathbf{u}',\lambda)\cdot\mathbf{S}(\mathbf{r},\mathbf{u},\lambda)+\mathbf{k}_{abs}(\mathbf{r},\lambda)\cdot\mathbf{S_0}(\mathbf{r},\mathbf{u},\lambda) \tag{1.1}$$

The spectral Stokes vector $\mathbf{S}(\mathbf{r},\mathbf{u},\lambda) = (I,Q,U,V)^T$ are the spectral VRTE solutions which encompass the spectral and polarimetric signatures of a particulate medium of interest as a function of position $\mathbf{r}$, direction $\mathbf{u}$ and wavelength λ. The phase matrix $P(\mathbf{r},\lambda)$ of an ensemble of particles is calculated by integrating the Mueller Matrix elements $M_{ij}(r_p,\mathbf{r},\lambda)$ over the particle size distribution $n(r_p)$ of an ensemble (i and j range from 1 to 4):

$$P(\mathbf{r},\lambda) = \begin{bmatrix} P_{11} & P_{12} & P_{13} & P_{14} \\ P_{21} & P_{22} & P_{23} & P_{24} \\ P_{31} & P_{32} & P_{33} & P_{34} \\ P_{41} & P_{42} & P_{43} & P_{44} \end{bmatrix} \tag{1.2}$$

with:

$$P_{ij}(\mathbf{r},\lambda) = \frac{\lambda}{\pi k_{sca}^2}\int dr_p M_{ij}(r_p,\mathbf{r},\lambda)n(r_p) \tag{1.3}$$

Particulate or scattering media are modeled for numerical purposes as semi-infinite plane-parallel layers with constant radiative parameters as shown in Fig. 1. For instance, a particulate medium inside a quartz or glass slab is modeled as a series of vertically inhomogeneous layers containing randomly oriented particles of various geometries. The general description of atmospheric layers is substituted herein by optical material interfaces, such as quartz or glass, with given complex refractive index. The extension of this model to spectral polarimetric light-scattering requires several modifications including spectral dispersion of the interfaces or solving Fresnel equations at every interface for every wavelength.

Our model based on the spectral VRTE solves numerically a system of L equations, where L is the number of discrete wavelengths composing the broadband light source (typically $L=1000$). The spectral operation consists of calculating the scattered Stokes vector corresponding at each wavelength in the spectral range. Azimuthal angles are expressed as a Fourier series and polarization rotations are performed directly in azimuth space. Subsequently, Fourier transform is performed to retrieve the scattering matrix for each Fourier azimuth mode [23,25,26]. Multiple reflections between layers are also taken into account in the spectral polarimetric radiance balance at the top and bottom of each layer. The adding–doubling technique is then deployed for each Fourier mode to model multiple scattering. This technique is numerically stable and is used to model multiple light scattering in an intuitive, efficient, and simple way. A good selection of the Fourier modes and the quadratic angles for every wavelength is crucial for the computation.

Validation of the model has been carried out following a three-step procedure [27]. Briefly, (i) an analytical validation was conducted for Rayleigh scattering from Coulson's table, (ii) a stochastic method was used to validate the model for Mie scattering and (iii) experiments were carried out on polystyrene particles in aqueous solution. As stated above, the spectral VRTE model is valid only for intermediate volume fractions, in dispersed-phase, as a large value of this parameter refers to a large number of multiple scattering events and multiple reflections inside a glass cuvette for instance.

3. Methodology

Spectral polarimetric light-scattering simulations are carried out using our spectral VRTE model for an incident broadband p-polarized collimated illumination. Typical radiative transfer inputs are optical thickness or albedo. However, for broadband calculations, these radiative parameters cannot be employed because of their spectral dependency. For broadband calculations, particulate media must be rather described in terms of microphysical parameters such as volume fraction or refractive index (both for particles and host medium), which are not dependent on the wavelength of illumination. Thus, a particulate scattering medium composed of an ensemble of random spherical particles is modeled with three principal microphysical parameters:

(1) Particle size distribution, which follows a log-normal size distribution such as

$$n(r_p) = \frac{dN(r_p)}{dr} = \frac{N_0}{\sqrt{2\pi}}\frac{1}{r_p\ln(\sigma)}\exp\left[-\frac{1}{2}\left(\frac{\ln(r_p/r_m)}{\ln(\sigma)}\right)^2\right] \tag{1.4}$$

where $n(r_p)$ is the particle size distribution as a function of the particle radius r_p, N_o is the total number density of particles, r_m is the mean particle radius and σ is the standard deviation of distribution.

(2) Complex optical index, which accounts for the spectral dispersion and absorption of both particles and host