

RETRIEVAL OF AEROSOL MICROPHYSICAL PROPERTIES FROM THE AERONET  
PHOTO-POLARIMETRIC MEASUREMENTS

by

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## CHAPTER 1

### INTRODUCTION

#### 1.1 Background and Motivation

Atmospheric aerosols play a crucial role in the global climate change. They affect earth energy budget directly by scattering and absorbing solar and terrestrial radiation, and indirectly through altering the cloud formation, lifetime, and radiative properties [Haywood and Boucher, 2000; Ramanathan *et al.*, 2001]. However, quantification of these effects in the current climate models is fraught with uncertainties. The global average of aerosol effective radiative forcing were estimated to range from  $-0.1$  to  $-1.9 \text{ Wm}^{-2}$  with the best estimate of  $-0.9 \text{ Wm}^{-2}$  [Boucher *et al.*, 2013], indicating that the cooling effects of aerosol might counteract the warming effects of  $1.82 \pm 0.19 \text{ Wm}^{-2}$  caused by the increase of carbon dioxide since the industrial revolution [Myhre *et al.*, 2013]. The climate effects of aerosol particles depend on their geographical distribution, optical properties, and efficiency as cloud condensation nuclei and ice nuclei. Key quantities pertain to the aerosol optical and cloud-forming properties include particle size distribution (PSD), chemical composition, mixing state, and morphology [Boucher *et al.*, 2013]. While the daily aerosol optical depth (AOD) can be well measured from current satellite and ground-based remote sensing instrumentations [e.g., Holben *et al.*, 1998; Kaufman *et al.*, 2002], the accurate quantification of aerosol ERF is in no small part hindered by our limited knowledge about the aerosol PSD and refractive index (describing chemical composition and mixing state).

To fully understand the role of aerosol particles in the global climate change, further development in observations along with retrieval algorithms for these aerosol microphysical properties from different platforms are thus highly needed [*Mishchenko et al.*, 2004], and the focus of this two-part series study is the characterization of aerosol properties from ground-based passive remote sensing.

### 1.1.1 Previous studies on aerosol microphysical retrievals

There have been continuous efforts in determining aerosol microphysical properties from ground-based measurements of direct and/or diffuse solar radiation since *Ångström* [1929] first suggested an empirical relationship between the spectral dependency of extinction coefficients and the size of aerosol particles. Over thirty years later, *Curcio* [1961] inferred the aerosol PSD from the spectral particulate extinction coefficients in the visible and near-infrared regions. Soon with the effective numerical inversion technique developed by *Phillips* [1962] and *Twomey* [1963] specifically for error-involved optimization, a number of studies explored the use of either spectral attenuations or scattered radiances (in a small range of scattering angles) to determine the aerosol PSD [*Twomey and Howell*, 1967; *Yamamoto and Tanaka*, 1969; *Dave*, 1971; *Grassl*, 1971; *Herman et al.*, 1971; *King et al.*, 1978]. *Shaw* [1979] and *Nakajima et al.* [1983] were among the first studies that have combined optical scattering measurements with spectral extinctions to recover particle size spectrum. *Kaufman et al.* [1994] suggested useful information contained in the sky radiances of larger scattering angles to retrieve the aerosol scattering phase function and PSD. The first operational retrieval algorithm for aerosol microphysical properties was introduced by *Nakajima et al.* [1996], when the multi-band automatic sun- and sky-scanning radiometer was deployed in the AErosol RObotic NETwork, or the AERONET [*Holben et al.*, 1998]. All of above mentioned methods treated aerosol particles as homogeneous



spheres and with refractive index assumed a priori, even though the refractive index can highly impact the optical, especially the scattering characteristics [*Hansen and Travis*, 1974]. *Tanaka et al.* [1982, 1983] developed an inversion library method to estimate the complex refractive index and PSD simultaneously from measurements of scattered radiances polarized in the perpendicular and parallel directions. Another concept for determining refractive index from both direct and diffuse angular radiances was developed by *Wendisch and Von Hoyningen-Huene* [1994] and *Yamasoe et al.* [1998], which were based on the fact that sensitivities of scattered radiances to the PSD and those to the refractive index are dominated on different scattering-angular regions. The current AERONET operational inversion algorithm was developed by *Dubovik and King* [2000], which has heritage from algorithms developed by *King et al.* [1978] and *Nakajima et al.* [1983, 1996] but was implemented for simultaneous retrieval of particle size distribution and complex refractive index with sophisticated inclusion of multiple a priori constraints. *Dubovik et al.* [2002a, 2006] further implemented the spheroids in the particle shape consideration for desert dust in the retrieval, and added fractional volume of non-spherical particles to the inversion products.

### 1.1.2 The AERONET measurements

With over 400 locations around the world, most AERONET sites are equipped with an automatic sun and sky scanning spectral radiometer, or the CIMEL-318 type SunPhotometer (Figure 1.1a), to routinely measure direct and diffuse solar radiation in various atmospheric window channels [*Holben et al.*, 1998]. As listed in Table 1.1 and illustrated in Figure 1.1, these measurements include direct sun radiances, sky radiance on both the solar almucantar and principal planes, as well as the optional polarization of sky light on the solar principal plane.

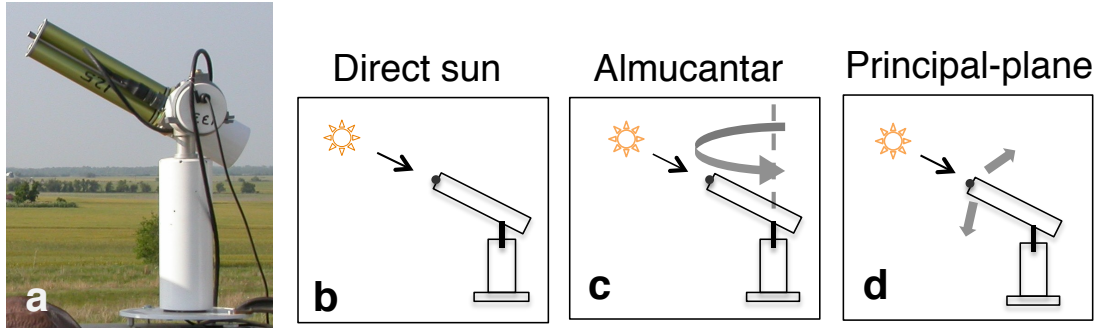


Figure 1.1: A photo of the CIMEL-318 type SunPhotometer and its observational modes.

Direct sun radiances at various atmospheric window channels from the ultra-violet (UV) to near-infrared (NIR) are used to infer the spectral AODs with the Beer-Lambert-Bouguer Law [Holben *et al.*, 1998; Smirnov *et al.*, 2000]. Depending on site-specific instruments, AOT values are typically reported at 7 wavelengths centered at 340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, and 1020 nm. Their calibration errors are believed to as small as 0.01 for visible and NIR bands and 0.02 for UV bands.

Sky radiance measurements, which are performed at 440, 670, 870, and 1020-nm bands with full width spectrum at half maximum (FWHM) of 10 nm, are acquired from both solar almucantar and solar principal plane. An almucantar is a series of measurements taken at the viewing angle of the sun for 76 specified relative azimuthal angles (for detail see table 2 of citeholben98). To achieve an enough range of scattering angles, almucantar scans are usually made at an optical air mass of 1.7 or more (corresponding to solar zenith angle larger than about  $50^\circ$ ). The principal-plane sequence for each spectrum performs right after almucantar scans. It begins with a sun observation, moves  $6^\circ$  below the sunray, sweeps up through the sun, and ends at a scattering angle of  $150^\circ$  or viewing angle achieves horizon, collecting radiances from up to 42 viewing angles. Hereinafter, we will use  $I_{\text{alm}}$  and  $I_{\text{ppl}}$  to represent the sky radiances from the solar almucantar and solar principal plane, respectively.

These sky radiance data are used in the current AEROENT operational inversion al-

Table 1.1: Measurement sequences of the CIMEL-318 SunPhotometer.

	Spectra (nm)	Viewing Geometry (°)	Applications
Direct sun	340–1020 340–1640 <sup>a</sup>	Target to the sun	AOD, $P_w$ , AE
Almucantar ( $I_{alm}$ )	440, 675, 870, 1020 (340, 380, 500, 1640) <sup>a</sup>	Azimuth angles relative to Sun: 6, 5, 4.5, 4, 3.5, 3, 2.5, 2, -2, -2.5, -3, -3.5, -4, -4.5, -5, -6, -8, -10, -12, -14, -16, -18, -20, -25, -30, -35, -40, -45, -50, -60, -70, -80, -90, -100, -110, -120, -130, -140, -160, -180 (Duplicate above sequence for a complete counter clockwise rotation to -6)	PSD, $m_r$ , $m_i$ , SSA, phase function
Principal- plane ( $I_{ppi}$ )	Same as above	Scattering angle from Sun: -6, -5, -4.5, -4, -3.5, -3, -2.5, -2, 2, 2.5, 3, 3.5, 4, 4.5, 5, 6, 8, 10, 12, 14, 16, 18, 20, 25, 30, 35, 40, 45, 50, 60, 70, 80, 90, 100, 110, 120, 130, 140 (negative is below the Sun)	Same as above
Polarization ( $I_{pp}$ , DOLP <sub>pp</sub> )	870, (340, 380, 440, 500, 675, 870, 1020, 1640) <sup>a</sup>	Zenith angle on the solar principal plane: -85, -80, -75, -70, -65, -60, -55, -50, -45, -40, -35, -30, -25, -20, -15, -10, -5, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85 (negative is in the antisolar direction)	Not used yet

<sup>a</sup>Additional measurements taken by the newer-generation CIMEL-318DP SunPhotometer.

gorithm [Dubovik and King, 2000; Dubovik et al., 2006] (hereafter Dubovik00&06) to derive: (1) the aerosol particle size distribution (PSD) in terms of the aerosol volume (in the atmospheric column) at 22 size bins, (2) the fractional volume of non-spherical particles, and (3) the complex refractive index assumed to be independent of particle size. From those microphysical parameters, the Dubovik00&06 algorithm computes the aerosol single scattering albedo (SSA) and the phase function. Uncertainties in the AERONET inversion products are 15–100% for the bin-based PSD parameters, 0.025–0.05 for real-part refractive index and 0.03 for SSA [Dubovik et al., 2000].

Light polarization measurements are performed optionally over many sites. They are measured by the SunPhotometer with three polarizers placed 60° between each axial direction. The total radiance is derived by

$$I_{pp} = \frac{2}{3} (I_1 + I_2 + I_3), \quad (1.1)$$

where  $I_1$ ,  $I_2$ , and  $I_3$  are radiance with these three polarizers, respectively. The degree of linear polarization (DOLP) of skylight is inferred by

$$\text{DOLP}_{\text{pp}} = \frac{2(I_1^2 + I_2^2 + I_3^2 - I_1 I_2 - I_2 I_3 - I_1 I_3)^{1/2}}{I_1 + I_2 + I_3}. \quad (1.2)$$

It should be noted that we prefer to use  $\text{DOLP}_{\text{pp}}$  instead of polarized radiance in our inversion, since as a relative quantity it is more accurate. Polarization measurements are made every hour (right after principal plane scans) at 870 nm in the principal plane at  $5^\circ$  increments between viewing zenith angle of  $-85^\circ$  and  $+85^\circ$ . These measurements are optional depending on the instrument version and configuration, and are currently available mostly over European and African stations. Recently, multi-spectral polarizations have also been taken with a newer-generation SunPhotometer (CIMEL CE318-DP) at some sites [[Li et al., 2009](#)] and the UAE<sup>2</sup> fields campaign [[Reid et al., 2008](#)]. Here we focus our study on using multi-spectral polarizations for the inversion of aerosol parameters.

### 1.1.3 Challenges and opportunities

While the AERONET AOD and other inversion products have been widely used to study the climatology of aerosol optical properties [[Dubovik et al., 2002b](#); [Levy et al., 2007a](#)] and for the development and validation of aerosol retrieval algorithms for satellite sensors such as the Moderate Resolution Imaging Spectrometer (MODIS) [[Kaufman et al., 1997](#); [Remer et al., 2005](#); [Levy et al., 2007b, 2010](#); [Wang et al., 2010](#)] and the Multi-angle Imaging SpectroRadiometer (MISR) [[Diner et al., 1998](#); [Kahn et al., 2010](#)], the AERONET operational algorithm also faces: (i) challenges in evaluation of aerosol data either retrieved from newer-generation satellite sensors or simulated from chemistry transport models, and (ii) opportunities to improve the retrieval through the use of multi-spectral polarization measurements that are now available at a few sites and will be made available at more sites as

part of the AERONET future research development (<http://aeronet.gsfc.nasa.gov>). These challenges and opportunities, as further described below, are also the motivation for us to develop a new research algorithm.

The first challenge is that newer-generation satellite sensors are expected to offer aerosol microphysical products with accuracy that is equivalent to, if not higher than, that of the current AERONET microphysical products. For instance, the Aerosol Polarimetry Sensor (APS) for the NASA Glory mission, through measuring the first three Stokes vector elements simultaneously from 250 viewing angles at nine spectral bands (410, 443, 556, 670, 865, 910, 1370, 1610, and 2200 nm), was designed to retrieve aerosol effective radius ( $r_{\text{eff}}$ ), effective variance ( $v_{\text{eff}}$ ), and spectral complex index of refraction for both fine and coarse modes [*Mishchenko et al.*, 2007]. While no actual product is available because of the failure of Glory launch, several case studies with the APS's prototype airborne sensor, RSP (the Remote Sensing Polarimeter), demonstrated feasibility of APS algorithm [*Chowdhary et al.*, 2002, 2005; *Mishchenko et al.*, 2004; *Waquet et al.*, 2009]. At least in the case of spherical particles, the accuracy of APS's bi-modal aerosol products was expected to be 10% for  $r_{\text{eff}}$ , 40% for  $v_{\text{eff}}$ , 0.02 for  $m_r$ , and 0.03 for the SSA ( $\omega_A$ ) [*Mishchenko et al.*, 2007]. Some of these accuracy expectations are unlikely to be matched by existing ground-based and in situ instruments, including those at the AERONET sites. Moreover, the current AERONET retrieval of the refractive index and the  $\omega_A$  are not recommended to use when the 440-nm AOD is lower than 0.4 [*Holben et al.*, 2006] due to expected limited accuracy identified in the detailed sensitivity study by [*Dubovik et al.*, 2000].

The second challenge is associated with the inconsistency in assumptions of PSD that exists between current AERONET inversion products and satellite retrievals on the one hand, as well as the aerosol models used by climate models on the other hand. Specifically, the Dubovik00&06 algorithm retrieves the aerosol PSD on in 22 discrete size bins. In contrast, a continuous PSD function (e.g., lognormal) is usually assumed in satellite retrieval

algorithms, such as those for APS/RSP [Mishchenko *et al.*, 2007; Waquet *et al.*, 2009] and the POLDER/PARASOL algorithm [Hasekamp *et al.*, 2011]. Also, aerosol microphysical properties are usually calculated with continuous PSD assumptions in many chemistry transport models, such as GEOS-Chem [Drury *et al.*, 2010; Wang *et al.*, 2010] and the GOCART model [Chin *et al.*, 2002]. Clearly, the actual aerosol PSD is never a perfect lognormal distribution, but neither it is discrete. At least from the scattering perspective, the aerosol PSD can be well characterized with an effective radius  $r_{\text{eff}}$  and an effective variance  $v_{\text{eff}}$ , while the specific function of the PSD is shown to be much less important [Hansen and Travis, 1974]. In other words, since the retrieval is based on the information content in the particle optical scattering, the most relevant size parameters, regardless of the PSD shape, should be  $r_{\text{eff}}$  and  $v_{\text{eff}}$ , at least for spherical particles.

The third challenge is that the assumption of a size-independent refractive index (and SSA) in Dubovik00&06 is not in line with the majority of counterpart satellite retrieval algorithms [e.g., Mishchenko *et al.*, 2007; Hasekamp *et al.*, 2011; Martonchik *et al.*, 2009], which often uses different refractive indices for various individual aerosol modes. In many cases, tropospheric aerosol is a mixture of modes with substantially different refractive indices. For example, smoke from biomass burning can be mixed with mineral dust over western coastal North Africa [Yang *et al.*, 2013]. Furthermore, the assumption of size-independent refractive index can lead to errors in the retrieval of the size distributions when the refractive indices for fine- and coarse-mode aerosols differ substantially [Dubovik *et al.*, 2000; Chowdhary *et al.*, 2001]. Thus, a mode-resolved parameterization of the refractive index in an aerosol retrieval algorithm not only can facilitate the validation of satellite products and chemistry transport models, but also is expected to improve the accuracy of PSD and SSA retrievals for each mode. [Dubovik *et al.*, 2000] have tested the possibility of retrieving separated refractive indices of fine and coarse modes, however, they concluded that the retrieval of bi-modal refractive indices is essentially non-unique due to limited

information in the AERONET radiance-only observations.

Therefore, this work aims to developing an algorithm to retrieve the aerosol microphysical properties of both fine and coarse aerosol modes, which embraces the future opportunities of deploying polarization measurements through AERONET, and ameliorates the aforementioned limitations in the Dubovik00&06 algorithm by incorporating both radiance and polarization data. Polarization measurements contain valuable information on aerosol microphysical properties [*Mishchenko and Travis, 1997*], as the polarization of the scattered light is highly sensitive to aerosol size and refractive index [*Hansen and Travis, 1974; Mishchenko et al., 2002*]. We note, however, their conclusions were based on consideration of spherical aerosol particles and were primarily from a theoretical point of view. In contrast, the studies by *Dubovik et al. [2006]* and *Deuzé et al. [1993, 2001]* revealed serious limitation of polarimetric retrieval of the properties for coarse, especially non-spherical aerosols. Moreover, *Dubovik et al. [2006]* have shown that while the polarimetric observation of fine particles and large spheres are highly sensitive to real part of refractive index, even they have non-negligible sensitivity to particle shape. Therefore, adding polarization measurements to the inversion has great potential to improve the accuracy of AERONET microphysical retrievals, provided that the difficulty of representing aerosol particle shapes is recognized or adequately addressed. In these regards, most of the past efforts seem to suggest clear improvements in characterization of fine mode aerosol using polarimetric observations. For example, *Li et al. [2009]*, based upon the Dubovik00&06 algorithm, demonstrated the possibility to reduce errors in the fine-mode size distribution, real part of the refractive index, and particle shape parameters.

## 1.2 Research Goals and Thesis Outline

As discussed above, this dissertation seeks to contribute to an improved research algorithm to retrieval aerosol micriphysical properties from AERONET measurements of light radiance and polarization, with emphasis on elucidating the potentially important role of polarization measurements. It does so by pursuing three following spcifci objectives:

1. *Develop ground-based inversion algorithms for the retrieval of  $r_{eff}$ ,  $v_{eff}$ ,  $m_r$ , and  $m_i$  from a combined use of direct solar radiance, skylight radiance and skylight linear polarization measurements from AERONET.*
2. *Perform a sensitivity study and error budgeting exercise to characterize retrieval accuracy and error sources.*

By doing so, we can answer the questions:

3. *Perform ground-based retrievals using available AERONET polarimetric measurements.*



## CHAPTER 2

### MODEL DEVELOPMENTS

#### 2.1 Introduction

The radiation fields—radiance and the state of polarization—measured by the AERONET SunPhotometer are the outcome of solar radiation interacting with various physical processes including the absorption and scattering by atmospheric molecules, aerosols and clouds, as well the reflection and absorption by underlying surface. The radiance and polarization of light at any wavelength can be represented by a Stokes column vector  $\mathbf{I}$  having four elements [*Hansen and Travis, 1974*]:

$$\mathbf{I} = [I, Q, U, V]^T, \quad (2.1)$$

where  $I$  is the total intensity (or radiance),  $Q$  and  $U$  describe the state of linear polarization,  $V$  describes the state of circular polarization, and  $T$  indicates a transposed matrix. It should be noted that all radiation fields and optical parameters used in this paper are functions of the light wavelength  $\lambda$ . For simplicity, however, we omit  $\lambda$  in all formulas. The degree of linear polarization (DOLP) is defined by

$$\text{DOLP} = \frac{\sqrt{Q^2 + U^2}}{I}. \quad (2.2)$$

In the solar principal plane,  $U$  is negligibly small and the above formula becomes  $\text{DOLP} = -Q/I$ . Let  $\mathbf{I}_0 = [I_0, 0, 0, 0]^T$  denote the Stokes vector for incident Solar radiation

at the top of the atmosphere (TOA) from the direction  $(\theta_0, \phi_0)$ , where  $\theta_0$  and  $\phi_0$  are the incident solar zenith and azimuth angles, respectively. For a plane-parallel atmosphere bounded below by a reflective surface, the vector radiative transfer equation in the medium for the specific intensity column vector  $\mathbf{I}$  of light propagating in the viewing direction  $(\theta, \phi)$  can be written [Hovenier *et al.*, 2004; Mishchenko *et al.*, 2002]:

$$\mu \frac{\partial \mathbf{I}(\tau, \mu, \phi)}{\partial \tau} = \mathbf{I}(\tau, \mu, \phi) - \mathbf{J}(\tau, \mu, \phi; \mu_0, \phi_0) \quad (2.3)$$

$$\begin{aligned} \mathbf{J}(\tau, \mu, \phi; \mu_0, \phi_0) = & \frac{\omega}{4\pi} \int_{-1}^1 \int_0^{2\pi} \mathbf{P}(\tau, \mu, \mu_0, \phi - \phi_0) \mathbf{I}(\tau, \mu_0, \phi_0) d\phi_0 d\mu_0 \\ & + \frac{\omega}{4\pi} \mathbf{P}(\tau, \mu, \mu_0, \phi - \phi_0) \mathbf{I}_0 \exp(-\tau/\mu_0) \end{aligned} \quad (2.4)$$

Here,  $\tau$  is the extinction optical depth measured from TOA,  $\mu$  and  $\mu_0$  are cosines of  $\theta$  and  $\theta_0$ , respectively,  $\omega$  is the SSA and  $\mathbf{P}$  is the phase matrix. The first term in equation (2.3) represents multiple scattering contributions, while the second indicates scattered light from the direct solar beam.

Parameters required to solve the above radiative transfer equation are  $\tau$ ,  $\omega$ , and  $\mathbf{P}(\Theta)$  for the atmosphere, and the reflectance matrix  $\mathbf{R}_s(\tau, \mu, \phi; \mu_0, \phi_0)$  of the underlying surface. Considering a cloud-free atmosphere, the solar radiation is attenuated by molecular scattering, gaseous absorption, and aerosol scattering and absorption. For a given layer, we have

$$\tau = \tau_A + \tau_R + \tau_G \quad (2.5)$$

$$\omega = \frac{\tau_A \omega_A + \tau_R}{\tau} \quad (2.6)$$

$$\mathbf{P}(\Theta) = \mathbf{P}_A(\Theta) \frac{\tau_A \omega_A}{\tau_A \omega_A + \tau_R} + \mathbf{P}_R(\Theta) \frac{\tau_R}{\tau_A \omega_A + \tau_R} \quad (2.7)$$

where  $\tau_A$ ,  $\tau_R$ , and  $\tau_G$  are optical depth, respectively, by aerosol extinction, Rayleigh scattering of air density fluctuations, and gaseous absorption.  $\omega_A$  is the SSA of aerosol, and

$\mathbf{P}_A(\Theta)$  and  $\mathbf{P}_R(\Theta)$  are, respectively, the aerosol and Rayleigh phase matrices as functions of the scattering angle  $\Theta$ . Therefore, the forward modeling development thus requires the computation of single scattering properties for aerosols and air density fluctuations, rigorous treatment for absorption of trace gases, accurate representation of reflectance/polarization by surface, and the realistic simulation of polarimetric radiative transfer.

In this regard, we have developed the UNified Linearized Vector Radiative Transfer Model, or UNL-VRTM, specifically for simulation, analysis, and inversion of the photo-polarimetric measurements. Components of the UNL-VRTM are described in section 2.2, and the model benchmarking and verification are presented in section 2.3.

## 2.2 The UNL-VRTM

As shown in Figure 2.1, the UNL-VRTM comprises 6 modules; they are

1. A module computing Rayleigh scattering (section 2.2.1);
2. A module that deal with gaseous absorption (section 2.2.1);
3. A linearized Mie scattering code (section 2.2.2);
4. A linearized T-matrix electromagnetic scattering code (section 2.2.2);
5. A surface model computing various bidirectional reflectance/polarization functions (BRDF/BPDF) (section 2.2.3);
6. A vector linearized radiative transfer model—VLIDORT (section 2.2.4).

These modules are integrated for the forward calculation of aerosol single scattering, gas absorption, and vector radiative transfer hereafter, and thus they together constitute the UNified Linearized Radiative Transfer Model, UNL-VRTM.

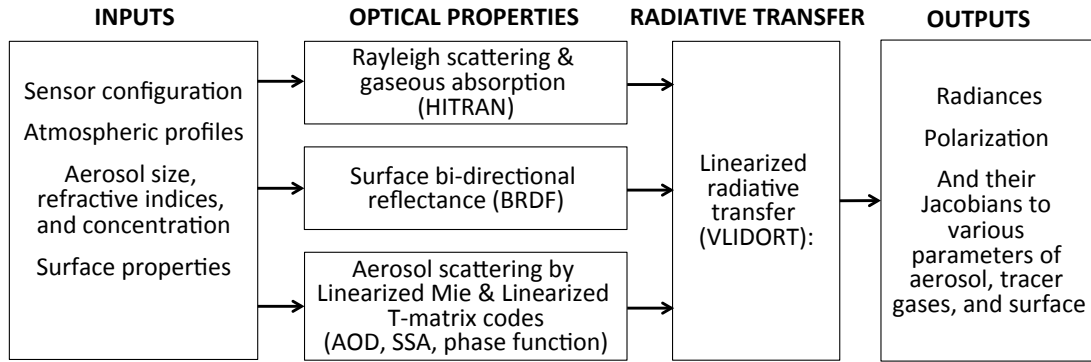


Figure 2.1: Flowchart of the UNL-VRM. See text for detail.

Inputs for the UNL-VRM are profiles of atmospheric properties and constituents (temperature, pressure, aerosol mass concentration or layer AOD, water vapor amount and other trace gas volume mixing ratio profiles), the surface properties, as well as the aerosol parameters (such as PSD parameters and refractive index) themselves. Bearing in mind the lack of sensitivity in passive remote sensing for the retrieval of vertical profiles of aerosol properties, the UNL-VRM as it stands now is only designed to deliver radiative calculations for a maximum of two sets of aerosol single scattering properties (e.g., aerosol PSD, refractive index, and particle shape), typically with one fine-mode and one coarse-mode aerosol. Other inputs for model include spectral and geometrical definitions that characterizing specification of an observing sensor.

Outputs of the model include the Stokes vector ( $\mathbf{I}$ ) at user-defined spectral wavelengths and desired atmospheric levels for both upwelling and downwelling radiation, from which the light radiance and degree of polarization can be derived. Outputs also include analytical Jacobians of  $\mathbf{I}$  with respect to all aerosol particle parameters (PSD, refractive index, vertical profile), Rayleigh scattering optical depth, optical depth of all trace gases, and parameters describing surface optical property. A detail description of the UNL-VRM's Jacobian capability is presented in section 2.2.5.

### 2.2.1 Molecular scattering and absorption

The Rayleigh scattering optical depth at certain wavelength in any atmospheric layer ( $\tau_R$ ) is computed by

$$\tau_R = N_{\text{air}} \sigma_R \quad (2.8)$$

where  $N_{\text{air}}$  is air molecular number density of that layer ( $\text{molec cm}^{-2}$ ), and  $\sigma_R$  is the Rayleigh scattering cross-section ( $\text{cm}^2 \text{ molec}^{-1}$ ) computed following [Bodhaine et al. \[1999\]](#). The Rayleigh phase matrix,  $\mathbf{P}_R(\Theta)$ , depends upon molecular anisotropy through the depolarization factor, also computed from the same source. [Bodhaine et al. \[1999\]](#) computes the wavelength-dependent Rayleigh scattering cross-section as a function of mixing ratios for  $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ , and  $\text{CO}_2$ . The phase matrix for Rayleigh scattering follows [Hansen and Travis \[1974\]](#); we use the set of spherical-function expansion coefficients for the phase matrix as supplied for VLIDORT [[Spurr, 2006](#)].

Calculation of the absorption optical depth ( $\tau_G$ ) at any atmospheric layer for  $K$  different trace gases follows

$$\tau_G = \sum_{i=1}^K N_{\text{gas},i} \sigma_{A,i}(T, P) \quad (2.9)$$

where  $N_{\text{gas},i}$  is the number density of  $i$ th gas within that layer, and  $\sigma_{A,i}$  is the corresponding absorption cross-section, a function of temperature and pressure. Our model accounts for absorptions by a total number of 22 trace gases:  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{O}_3$ ,  $\text{N}_2\text{O}$ ,  $\text{CO}$ ,  $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{NO}$ ,  $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{NH}_3$ ,  $\text{HNO}_3$ ,  $\text{OH}$ ,  $\text{HF}$ ,  $\text{KCl}$ ,  $\text{HBr}$ ,  $\text{HI}$ ,  $\text{ClO}$ ,  $\text{OCS}$ ,  $\text{H}_2\text{CO}$ ,  $\text{HOCl}$ , and  $\text{N}_2$ . The determination of  $\sigma_A$  utilizes a UV-to-visible cross-section library and the line-spectroscopic absorption parameters archived in the HITRAN database [[Orphal and Chance, 2003](#); [Rothman et al., 2009](#)]. The cross-section library compiles the extinction cross-section for  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{SO}_2$ , and  $\text{O}_2\text{--O}_2$  in the UV and/or visible spectral regions. Meanwhile, line-spectroscopic absorption databased are used to simulate the pressure- and temperature-dependent extinc-

tion cross-section with line-by-line (LBL) approach [Liou, 2002; Rothman *et al.*, 2009] by accumulating each individual absorption line. Doppler broadening is calculated from the molecular mass and the temperature, and Doppler and Lorentz broadening are included in the Voigt calculation.

Particular to work, we only consider the most influential trace species for the AERONET spectral bands: H<sub>2</sub>O (vapor), O<sub>3</sub>, and NO<sub>2</sub>. In our algorithm (section 3), the columnar amounts of O<sub>3</sub> and NO<sub>2</sub> are dynamically adjusted with retrievals from the Ozone Monitoring Instrument (OMI) [Levelt *et al.*, 2006] on board the AURA satellite. We apply the columnar water vapor amount retrieved from the 940-nm radiances measured by the AERONET SunPhotometer [Halthore *et al.* [1997].

### 2.2.2 Aerosol single scattering

Aerosol single scattering properties necessary to the radiative transfer calculation include aerosol optical depth ( $\tau_A$ ) ( $Q_{\text{ext}}$ ), SSA ( $\omega_A$ ), and scattering phase matrix ( $\mathbf{P}_A(\Theta)$ ). The calculation of these parameters is made with a Linearized Mie (LMIE) scattering electromagnetic code for spherical particles and a Linearized T-matrix (LTMATRIX) scattering code for non-spherical convex and axially symmetric particles [Spurr *et al.*, 2012]. The LMIE code originates from the Mie code of *de Rooij and Stap* [1984], and the LTMATRIX code originates from the T-Matrix code developed by *Mishchenko et al.* [1996]; *Mishchenko and Travis* [1998]; both include linearization capability developed by *Spurr et al.* [2012].

Common inputs for both codes are the complex refractive index ( $m_r + im_i$ ), and the particle size distribution (PSD) parameters for polydisperse scattering. The codes have several options to specify the PSD function: two-parameter gamma, two-parameter lognormal, three-parameter modified gamma, and four-parameter bi-lognormal. In addition, the linearized T-matrix code offers options to characterize the shape of non-spherical aerosols

(spheroids, cylinders, or Chebyshev particles) [Spurr *et al.*, 2012]. For non-spherical particles, the specified size distribution is interpreted as the equivalent surface-area sphere in the linearized T-matrix calculation, regardless of the shape.

For AERONET inversion algorithm, we assume that the aerosol volume distribution follows a bi-modal lognormal function [in agreement with Schuster *et al.*, 2006; Waquet *et al.*, 2009]:

$$\frac{dV}{d \ln r} = \sum_{i=1}^2 \frac{V_0^i}{\sqrt{2\pi \ln \sigma_g^i}} \exp \left[ -\frac{(\ln r - \ln r_v^i)^2}{2 \ln^2 \sigma_g^i} \right] \quad (2.10)$$

where  $V_0$ ,  $r_v$ , and  $\sigma_g$  are the total volume concentration, geometric median radius, and standard deviation, respectively. The superscript  $i$  indicates the size mode, and later will be replaced by ‘f’ for fine mode and ‘c’ for coarse mode. We assume that particle size ranges from 0.01 to 10  $\mu\text{m}$  for the fine mode and from 0.05 to 20  $\mu\text{m}$  for the coarse mode, both covering  $> 99.9\%$  of the total volume of an idealistic size range  $(0, +\infty)$ . An advantage of the lognormal distribution is that standard deviations for the number, area, and volume PSD functions are identical, and therefore allowing that the median radii for these PSD functions can be converted from one to another [Seinfeld and Pandis, 2006]. The  $r_{\text{eff}}$  and  $v_{\text{eff}}$  are related to the geometric parameters through:

$$r_{\text{eff}} = r_v \exp \left( -\frac{1}{2} \ln^2 \sigma_g \right), \quad (2.11)$$

$$v_{\text{eff}} = \exp (\ln^2 \sigma_g) - 1. \quad (2.12)$$

The LMIE/LTMATRIX code computes the aerosol extinction efficiency factor  $Q_{\text{ext}}$ , single scattering albedo  $\omega_A$ , and phase matrix  $\mathbf{P}_A(\Theta)$ , as well as Jacobians of these quantities with respect to input parameters including  $r_{\text{eff}}$ ,  $v_{\text{eff}}$ ,  $m_r$ , and  $m_i$ . The phase matrix and its Jacobians are expressed in terms of the coefficients  $\mathbf{B}_A(\Theta)$  for each moment  $l$  in terms of the generalized spherical function expansions for each non-zero phase matrix element. Let

$\mathbf{A}$  denotes the vector of aerosol microphysical parameters,  $\mathbf{A} = [V_0, r_{\text{eff}}, v_{\text{eff}}, m_{\text{r}}, m_{\text{i}}]^T$ , and  $\mathbf{M}$  the vector of aerosol optical parameters,  $\mathbf{M} = [\tau_{\text{A}}, \omega_{\text{A}}, \mathbf{B}_{\text{A}}(\Theta)]^T$ , where  $\tau_{\text{A}}$  is related to  $Q_{\text{ext}}$  by  $\tau_{\text{A}} = \frac{3V_0 Q_{\text{ext}}}{4r_{\text{eff}}}$ . The LMIE/LTMATRIX code acts as an operator that maps vector  $\mathbf{A}$  to  $\mathbf{M}$ . The Jacobian matrix of  $\mathbf{M}$  with respect to  $\mathbf{A}$  calculated by means of the linearization feature of the code, and it can be expressed by  $\nabla_{\mathbf{A}}\mathbf{M}$ .

### 2.2.3 Surface representations

VLIDORT has a supplementary module for specification of the surface BRDF as a linear combination of (up to) three semi-empirical kernel functions; for details, see [Spurr \[2004\]](#). This supplementary module can also provide partial derivatives of the BRDF with respect to the kernel weighting factors or with respect to kernel parameters such as the wind speed for glitter reflectance. These kernel functions include Lambertian, Ross-Thick, and Li-Sparse functions [[Wanner et al., 1995](#); [Lucht et al., 2000](#)], a Bi-directional Polarization Distribution Function (BPDF) [[Maignan et al., 2009](#)], and an ocean surface model based on the Cox-Munk model [[Cox and Munk, 1954](#)]. In addition, VLIDORT has an option for using a surface-leaving radiation field, either as a fluorescence term or as a water-leaving term expressed as a function of chlorophyll absorption.

Although surface reflectance has in general a low influence on AERONET down-welling sky radiances and polarization, a state-of-the-art representation of the surface reflectivity potentially reduces model uncertainties, especially for measurements taken at low elevation angles that could be affected by surface diffusion. Here, we utilize the spectral BRDF parameters from the MODIS surface products that are operationally reported every 16 days at a 1-km resolution [[Lucht et al., 2000](#)]. Here we use time-matched MODIS BRDF products to reconstruct the bidirectional reflectance over AERONET stations. The MODIS BRDF product supplies three weighting parameters ( $f_{\text{iso}}$ ,  $f_{\text{vol}}$ , and  $f_{\text{geo}}$ ) for the first 7 MODIS



bands, respectively, corresponding to three kernel types: isotropic, Ross-Thick ( $K_{\text{vol}}$ ), and Li-Sparse ( $K_{\text{geo}}$ ):

$$\rho_{\text{R}}(\mu, \phi; \mu_0, \phi_0) = f_{\text{iso}} + f_{\text{vol}}K_{\text{vol}}(\mu, \phi; \mu_0, \phi_0) + f_{\text{geo}}K_{\text{geo}}(\mu, \phi; \mu_0, \phi_0) \quad (2.13)$$

Expanded expressions for  $K_{\text{vol}}$  and  $K_{\text{geo}}$  appear in [Wanner et al. \[1995\]](#); [Lucht et al. \[2000\]](#).

Studies have shown that the BPDF for land surfaces is generally rather small and is “spectrally neutral” [[Nadal and Breon, 1999](#); [Maignan et al., 2004, 2009](#); [Waquet et al., 2007](#); [Litvinov et al., 2011](#)]. Most empirical BPDF models are based on Fresnel coefficients of light reflectance from the surface. Here we have incorporated the one-parameter model developed by [Maignan et al. \[2009\]](#), which was derived from analyses of several years of POLDER/PARASOL measurements. This model describes the polarized reflectance at any viewing geometry  $(\mu, \phi)$  from the given incident geometry  $(\mu_0, \phi_0)$  as:

$$\rho_{\text{P}}(\mu, \phi; \mu_0, \phi_0) = \frac{C_0 \exp(-\tan \theta_{\text{h}}) \exp(-\text{NDVI})}{\mu_0 + \mu} \mathbf{F}_{\text{P}}(\theta_{\text{h}}, n_{\text{v}}) \quad (2.14)$$

where  $C_0$  is a constant parameter chosen for a certain surface type,  $\theta_{\text{h}}$  is half of the phase angle of reflectance,  $n_{\text{v}}$  is the refractive index of vegetation (1.5 is used), and  $\mathbf{F}_{\text{P}}$  is the Fresnel reflection matrix. We chose a spectrally-independent value for  $C_0$  based on the recommendations by [Maignan et al. \[2009\]](#) for relevant surface types.

The combination of the BRDF and BPDF for land surface follows the discussion by [Dubovik et al. \[2011\]](#). The surface reflectance matrix  $\mathbf{R}_{\text{s}}(\mu, \phi; \mu_0, \phi_0)$  is represented as a sum of diffuse unpolarized reflectance and specular reflectance; the former is modeled using the MODIS BRDF in equation (2.13), and the latter using the BPDF formula in equation (2.14).

### 2.2.4 Radiative transfer

The radiative transfer equation (2.3) is solved with the Vector Linearized Discrete Ordinate Radiative Transfer (VLIDORT) model, which is a core part of the UNL-VRM. VLIDORT, developed by *Spurr* [2006], is a linearized pseudo-spherical vector discrete ordinate radiative transfer model for multiple scattering of diffuse radiation in a stratified multi-layer atmosphere. It computes four elements of the Stokes vector  $\mathbf{I}$  for downwelling and upwelling radiation at any desired atmospheric level. The VLIDORT includes the pseudo-spherical approximation to calculate solar beam attenuation in a curved medium. It also uses the delta-M approximation for dealing with sharply peaked forward scattering. Specifically for the AERONET inversion, we consider 16 discrete ordinate streams in the radiative transfer calculation and retain 180 terms in the spherical-function expansion of the scattering matrix to ensure accurate calculation of diffuse radiation.

Along with the Stokes vector  $\mathbf{I}$ , VLIDORT also computes the Jacobian matrix of  $\mathbf{I}$  with respect to aerosol optical vector  $\mathbf{M}$ ,  $\nabla_{\mathbf{M}}\mathbf{I}$ . Therefore, the combination of the VLIDORT and the LMIE/LTMATRIX codes allows for a direct calculation of the Jacobian matrix of the Stokes vector with respect to aerosol microphysics,  $\mathbf{A}$ , by

$$\nabla_{\mathbf{A}}\mathbf{I} = \nabla_{\mathbf{M}}\mathbf{I} \cdot \nabla_{\mathbf{A}}\mathbf{M} \quad (2.15)$$

Essentially, the above equation can yield the derivatives of the radiance  $I$  and DOLP with respect to any aerosol microphysical parameter, i.e.,  $\nabla_{\mathbf{A}}I$  and  $\nabla_{\mathbf{A}}\text{DOLP}$ . While obtaining  $\nabla_{\mathbf{A}}I$  is straightforward,  $\nabla_{\mathbf{A}}\text{DOLP}$  can be derived from equation (2.2) following:

$$\nabla_{\mathbf{A}}\text{DOLP} = -\frac{\text{DOLP}\nabla_{\mathbf{A}}}{I} + \frac{Q\nabla_{\mathbf{A}}Q + U\nabla_{\mathbf{A}}U}{I\sqrt{Q^2 + U^2}} \quad (2.16)$$

Table 2.1: Elements of transformation vector for various aerosol single scattering parameters (composite of fine and coarse mode).

$x$	$\phi_x$	$\varphi_x$	$\Psi_x^j$
$\tau_A$	$\frac{\tau_A}{\tau}$	$\frac{\tau_A}{\tau} \left( \frac{\omega_A}{\omega} - 1 \right)$	$\begin{cases} \frac{\omega_A \tau_A}{\omega \tau} \left( \frac{\mathbf{B}_A^j}{\mathbf{B}^j} - 1 \right) & \text{for } j < 3 \\ \frac{\tau_R}{\omega \tau} & \text{for } j \geq 3 \end{cases}$
$\omega_A$	0	$\frac{\tau_A \omega_A}{\tau \tau_A \omega_A + \tau_R}$	Same as above
$B_A^j$	0	0	$\begin{cases} \frac{\omega_A \tau_A \mathbf{B}_A^j}{\omega_A \tau_A \mathbf{B}_A^j + \tau_R \mathbf{B}_R^j} & \text{for } m = j < 3 \\ 1 & \text{for } m = j \geq 3 \\ 0 & \text{for } m \neq j \end{cases}$

### 2.2.5 Capability of calculating Jacobians

This section analytically derives the Jacobian of  $\mathbf{I}$  with respect to various aerosol related parameters, including  $\tau_A$ ,  $\omega_A$ ,  $\mathbf{B}_A$ , refractive index, PSD parameters, and aerosol vertical profile.

## 2.3 Model Benchmarking and Verifications

## CHAPTER 3

### INVERSION ALGORITHM

#### **3.1 General sturcture**

Basic formulation of inverse problem; sturcture of the algorithm.

#### **3.2 Combine a priori and smoothness constraints**

#### **3.3 Statistical optimized inversion**

#### **3.4 Retrieval Error Characertization**

#### **3.5 Qaulity Control of Measurements**

## CHAPTER 4

### INFORMATION CONTENT ANALYSIS

#### **4.1 Introduction**

#### **4.2 Inversion and Information Theories**

##### **4.2.1 Inverse problem and error characterization**

#### **4.3 Experiment Design**

#### **4.4 Results**

##### **4.4.1 Error-normalized (EN) Jacobian matrix**

##### **4.4.2 Information content and retrieval error**

###### **4.4.2.1 Aerosol PSD**

###### **4.4.2.2 Refractive indices**

###### **4.4.2.3 Single scattering albedo**

To be filled ...

## **4.5 Sensitivity of retrieval error to AOD and fmf**

To be filled ...

## **4.6 Summary**

To be filled ...

## CHAPTER 5

### CASE DEMONSTRATIONS

**5.1 Selected case and the a priori characterization**

**5.2 Fitting Residuals**

**5.3 Retrieved Aerosol Properties**

**5.4 Improvement over Radiance-Only Retrievals**

**5.5 Summary**

## CHAPTER 6

### SUMMARY AND OUTLOOK

#### **6.1 Summary of the Dissertation**

#### **6.2 Main Conclusions of This Work**

#### **6.3 Outlook and Future Work**



APPENDIX A

**ABBREVIATIONS AND ACRONYMS**

## APPENDIX B

### **SYMBOLS**

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