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

by

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Table of Contents

List of Figures

List of Tables

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 Wm^{-2} with the best estimate of –0.9 Wm^{-2} [*Boucher et al.*, 2013], indicating that the cooling effects of aerosol might counteract the warming effects of 1.82±0.19 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 word, 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 XX as illustrated in Figure 1.1, these measurements include direct sun radiances, sky radiance on both the solar almuncantar and pruncipal planes, as well as the optional polarization of sky light on the solar principal plane.

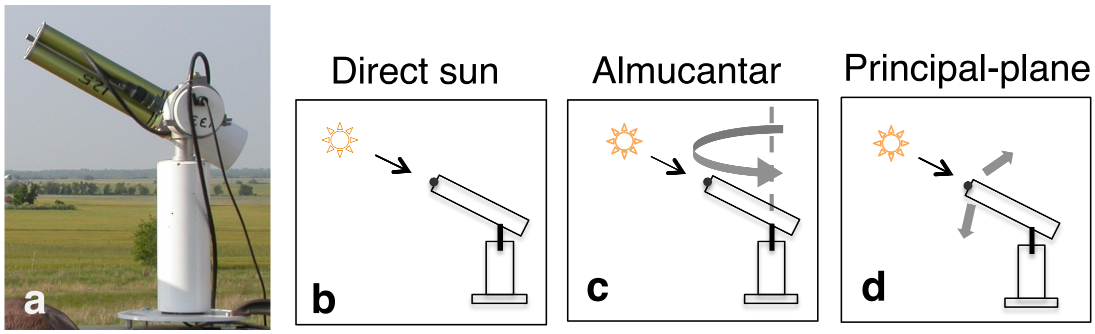

Figure 1.1: A photo of the CIMEL-318 type SunPhotometer and its observational modes.

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 citepholben98). 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°). The principal-plane sequence for each spectrum performs right after almucantar scans. It begins with a sun observation, moves 6°) below the sunray, sweeps up through the sun, and ends at a scattering angle of 150°) or viewing angle achieves horizon, collecting radiances from up to 42 viewing angles. Hereinafter, we will use I_{alm} and I_{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 algorithm [*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&06algorithm 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

$$I_{\rm pp} = \frac{2}{3}\left(I_1 + I_2 + I_3\right), \tag{1.1}$$

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

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

It should be noted that we prefer to use $\mathrm{DOLP}_{\rm pp}$ 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° increments between viewing zenith angle of $-85°$ and $+85°$. 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^2 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_{eff}), effective variance (v_{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_{eff}, 40% for v_{eff}, 0.02 for m_{r}, and 0.03 for the SSA (ω_{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 ω_{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&06algorithm 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_{eff} and an effective variance v_{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_{eff} and v_{eff}, at least for spherical particles.

The third challenge is that the assumption of a size-independent refractive index (and SSA) in Dubovik00&06is 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.

1.2 Objectives

1.3 Organization

CHAPTER 2

MODEL DEVELOPMENTS

2.1 Introduction

The radiation fields—radiance and the state of polarization—measured by the remote sensing instruments are the outcome of the interations of solar radiation 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

2.2 The UNL-VRTM

2.2.1 Surface representations

2.2.2 Molecular scattering and absorption

2.2.3 Aerosol single scattering

2.2.4 Radiative transfer

2.2.5 Capability of calculating Jacobians

2.3 Model Benchmarking and Verifications

CHAPTER 3

INFORMATION CONTENT ANALYSIS

3.1 Introduction

3.2 Inversion and Information Theories

3.2.1 Inverse problem and error characterization

3.3 Experiment Design

3.4 Results

3.4.1 Error-normalized (EN) Jacobian matrix

3.4.2 Information content and retrieval error

3.4.2.1 Aerosol PSD

3.4.2.2 Refractive indices

3.4.2.3 Single scattering albedo

To be filled . . .

3.5 Sensitivity of retrieval error to AOD and fmf

To be filled . . .

3.6 Summary

To be filled . . .

CHAPTER 4

INVERSION ALGORITHM

4.1 General sturcture

Basic formulation of inverse problem; sturcture of the algorithm.

4.2 Combine a priori and smoothness constraints

4.3 Statistical optimized inversion

4.4 Retrieval Error Characertization

4.5 Qaulity Control of Measurements

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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