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Validation of LIRIC aerosol concentration retrievals using airborne measurements during a biomass burning episode over Athens

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ABSTRACT

In this paper we validate the Lidar-Radiometer Inversion Code (LIRIC) retrievals of the aerosol concentration in the fine mode, using the airborne aerosol chemical composition dataset obtained over the Greater Athens Area (GAA) in Greece, during the ACEMED campaign. The study focuses on the 2nd of September 2011, when a long-range transported smoke layer was observed in the free troposphere over Greece, in the height range from 2 to 3 km. CIMEL sun-photometric measurements revealed high AOD (~0.4 at 532 nm) and Ångström exponent values (~ 1.7 at 440/870 nm), in agreement with coincident ground-based lidar observations. Airborne chemical composition measurements performed over the GAA, revealed increased CO volume concentration (~ 110 ppbv), with 57% sulphate dominance in the PM_1 fraction. For this case, we compare LIRIC retrievals of the aerosol concentration in the fine mode with the airborne Aerosol Mass Spectrometer (AMS) and Passive Cavity Aerosol Spectrometer Probe (PCASP) measurements. Our analysis shows that the remote sensing retrievals are in a good agreement with the measured airborne in-situ data from 2 to 4 km. The discrepancies observed between LIRIC and airborne measurements at the lower troposphere (below 2 km), could be explained by the spatial and temporal variability of the aerosol load within the area where the airborne data were averaged along with the different time windows of the retrievals.

1. Introduction

According to the report of the Intergovernmental Panel on Climate Change (IPCC; Myhre et al., 2013), anthropogenic aerosols result into a net cooling globally through their interaction with radiation and clouds, by an amount that remains difficult to quantify accurately and which could be comparable in magnitude to the net warming effect of greenhouse gases. Moreover, according to the World Health Organization (WHO), there is a significant linkage between suspended particles and human's mortality (WHO, 2006). Aerosol effects are determined, among others, by particle's size, their chemical composition, and number concentration. To understand how atmospheric particles are affecting the Earth's climate, the scientific community has established and operates global networks equipped with active and passive remote

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sensing instrumentation. More precisely, the AErosol RObotic NETwork (AERONET) provides almost real-time columnar aerosol optical and microphysical properties, based on the operation of >400 sun-sky radiometers distributed worldwide (Holben et al., 1998). Several aerosol lidar networks are used for aerosol/cloud research including well established infrastructures and networks like: the European Aerosol Research Lidar Network (EARLINET; Pappalardo et al., 2014), the Micro Pulse Lidar Network (MPLNET; Welton and Campbell, 2002) and the Asian Dust and aerosol lidar observation network (AD-Net; Sugimoto et al., 2014).

Depending on the capabilities of each system and the employed techniques, lidar measurements are used to retrieve the vertical distribution of (a) the aerosol backscatter coefficient (β_{aer}), (e.g. Fernald et al., 1972; Klett, 1981); (b) the aerosol extinction coefficient (a_{aer}), (Ansmann et al., 1990; Ansmann et al., 1992) and (c) the particle linear depolarization ratio δ_{aer} (e.g. Cairo et al., 1999; Sassen, 2005; Freudenthaler et al., 2009). The aerosol backscatter and extinction coefficients can be retrieved from the backscatter, Raman (Ansmann et al., 1992; Whiteman et al., 1992; Whiteman, 2003), or High Spectral Resolution Lidar (HSRL) technique (e.g. Eloranta, 2005). The selected technique defines also the accuracy of the retrieved aerosol products. Many studies have demonstrated that the spectral information of the aforementioned aerosol optical properties, makes feasible the provision of accurate retrievals in the fine mode, regarding aerosol microphysical parameters namely: aerosol size distribution, aerosol effective radius, number and volume concentration (Müller et al., 1999; Veselovskii et al., 2002; Veselovskii et al., 2010). In case that the aerosol extinction coefficient is provided by Raman technique, the microphysical retrievals are typically limited to night-time measurements since their accuracy depends on the error of the optical parameters provided as initial inputs.

During the last years, there are increased efforts to retrieve aerosol concentration profiles also during day-time. For example, the polarization lidar photometer networking (POLIPHON) technique is capable of retrieving the concentration profiles of dust and non-dust particles using single wavelength backscatter and depolarization coefficient lidar profiles (Ansmann et al., 2011; Ansmann et al., 2012). In this technique, AERONET microphysical retrievals are used to provide columnar volume-to-AOD values needed to convert the optical properties to concentration. On a continuation effort, Mamouri and Ansmann (2014), have expanded the technique to separating the contribution of fine and coarse dust modes, based on laboratory measurements of fine and coarse dust depolarization ratios.

In the framework of Aerosols, Clouds, and Trace gases Research Infrastructure (ACTRIS), two algorithms have been developed for retrieving concentration profiles from the synergy of lidar and sun-photometric measurements. The Generalized Aerosol Retrieval from Radiometer and Lidar Combined (GARRLiC; Lopatin et al., 2013) inversion algorithm retrieves vertical profiles of both fine and coarse aerosol concentrations as well as the size distribution and complex refractive index for each mode. Based on similar approach, the LIdar-Radiometer Inversion Code (LIRIC; Chaikovsky et al., 2016) considers that the fine and coarse particle intensive properties are constant with height, taken equal to the column-integrated values provided by AERONET, with only their concentration varying along the atmospheric column. LIRIC, GARRLiC and POLIPHON techniques have been used by many EARLINET-AERONET stations, during large and medium scale dust events over the European continent, for evaluating dust model performance in terms of dust layer geometrical properties (height range and centre of mass) as well as dust load (particle concentrations) (e.g. Binietoglou et al., 2015; Granados-Muñoz et al., 2016a, 2016b). For a case study of Saharan dust outbreak over Athens, Greece, Tsekeri et al. (2013), found a satisfactory agreement between LIRIC output and dust concentration profiles, simulated by the regional dust model BSC-DREAM8b (Pérez et al., 2006a; Pérez et al., 2006b; Basart et al., 2012). Furthermore, comparisons of LIRIC output, with dispersion models of other aerosol types than dust (e.g. volcanic dust), like Lagrangian dispersion model FLEXPART (Stohl et al., 1998; Stohl et al., 2005), showed a Pearson's coefficient (R) varying from 0.69 to 0.84 (Kokkalis et al., 2013). Moreover, Wagner et al. (2013), inter-compared LIRIC and POLIPHON concentration profiles, by applying both techniques on two case studies of irregularly shaped particles in the atmosphere (i.e. one Sahara dust outbreak and one volcanic dust event). The comparison between the two techniques revealed acceptable agreement, however the potential of LIRIC to retrieve optical properties, namely particle backscatter coefficient, lidar ratio and Ångström exponent, was found to demonstrate systematic deviations compared to corresponding measurements obtained with a Raman lidar. In addition, Papayannis et al. (2014) showed that the relative difference between LIRIC and POLIPHON mass concentration retrievals, is in the range of ±20% for the case of coarse non-spherical particles.

Furthermore, LIRIC retrievals of volume concentration have been compared with aircraft in-situ measurements, by Granados-Muñoz et al. (2016a) during a Saharan dust episode over Granada, Spain. The case study of coarse mode (dust), non-spherical particles, is introducing limitations regarding, the potential of in-situ instrumentation to measure size distribution in the size range above 3 μm (diameter). Thus, during their study, they combined: in-situ depolarization measurements from Cloud and Aerosol Spectrometer with Polarization detection (CAS-POL; Baumgardner et al., 2001), operating at the size range 0.6–50 μm (diameter), and a Passive Cavity Aerosol Spectrometer (PCASP 100×; Rosenberg et al., 2012; Cai et al., 2013) measuring aerosol size distribution in 0.1–3 μm diameter range. Correcting the retrieved size distributions, for refractive index assumptions, they demonstrated volume concentration discrepancies <20 μm^3 cm^{-3} (0.02 ppbv) and they attributed them to CAS-POL overestimation due to the asphericity of dust particles and to the possible underestimation of LIRIC, despite the fact that the derived size distributions from CAS-POL and lidar, were found to be in a good agreement.

In this study, we validate the volume concentration retrieved by LIRIC, with independent in-situ measurements of chemical composition, for a case of predominant fine mode particles in the atmosphere, over the Greater Athens Area (GAA; Saronic Gulf, Evoikos Gulf and Aegean), Greece. To our knowledge, this is the first time that fine mode LIRIC retrievals are validated against airborne. The case of fine mode particles is favourable for comparing remote sensing and in-situ observations since in this case there are fewer limitations on the instrumental side for the in-situ measurements while for remote sensing part the Mie scattering simulations are applicable since the particles are mainly spherical. In Section 2 we present the instrumentation and methodology used. Section 3 presents a brief description of ACEMED campaign along with the case study used for the evaluation of the LIRIC fine mode aerosol concertation retrieval. Our analysis contains, a thorough characterization of the aerosol load monitored over the GAA, in terms of their optical properties and chemical composition. In the second part of Section 3 we compare the retrieved concentrations with the independent in-situ airborne measurements. Finally, our conclusions are given in Section 4.

2. Instrumentation and method

2.1. Backscatter-depolarization lidar

At the National Technical University of Athens (NTUA, 37.97° N, 23.79° E, elevation: 212 m) a six-wavelength Raman-backscatter lidar system (EOLE) operates since February 2000, as a member of the EARLINET network (Bösenberg et al., 2003; Pappalardo et al., 2014). The emission unit is based on an Nd:YAG laser, emitting high energy laser pulses at 355, 532 and 1064 nm with a repetition rate of 10 Hz. The respective emitted energies per pulse are of the order of 240, 300 and 260 mJ. A Galilean type beam expander (×3) is mounted, just before the emission of the laser beam in the atmosphere, for reducing the laser beam divergence and increasing the beam diameter, almost