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Using Raman-lidar-based regularized microphysical retrievals and Aerosol Mass Spectrometer measurements for the characterization of biomass burning aerosols

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ABSTRACT

In this work we extract the microphysical properties of aerosols for a collection of measurement cases with low volume depolarization ratio originating from fire sources captured by the Raman lidar located at the National Institute of Optoelectronics (INOE) in Bucharest. Our algorithm was tested not only for pure smoke but also for mixed smoke and urban aerosols of variable age and growth. Applying a sensitivity analysis on initial parameter settings of our retrieval code was proved vital for producing semi-automatized retrievals with a hybrid regularization method developed at the Institute of Mathematics of Potsdam University. A direct quantitative comparison of the retrieved microphysical properties with measurements from a Compact Time of Flight Aerosol Mass Spectrometer (CToF-AMS) is used to validate our algorithm. Microphysical retrievals performed with sun photometer data are also used to explore our results. Focusing on the fine mode we observed remarkable similarities between the retrieved size distribution and the one measured by the AMS. More complicated atmospheric structures and the factor of absorption appear to depend more on particle radius being subject to variation. A good correlation was found between the aerosol effective radius and particle age, using the ratio of lidar ratios (LR: aerosol extinction to backscatter ratios) as an indicator for the latter. Finally, the dependence on relative humidity of aerosol effective radii measured on the ground and within the layers aloft show similar patterns.

1. Introduction

Biomass burning aerosols (BBA) are a key component of earth's atmosphere, and yet their exact effect on climate is poorly understood [34]. BBA are known to affect the climate both by scattering and absorbing solar radiation and by modifying the properties of clouds, but the exact magnitude of these effects depends both on their content in black carbon and their vertical distribution. For example, Brioude et al. [17] have shown that BBA can both increase and decrease stratocumulus

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cloud cover above oceans, depending on whether they are located above or within the marine boundary layer, respectively. Depending on their altitude, aerosol layers containing black carbon can alter the planetary albedo by absorbing the radiation scattered by low clouds, and accordingly change the cloud formation by heating the lower part of the atmosphere, causing clouds to evaporate faster [56]. At the same time, non-absorbing aerosol in the biomass burning plumes can have the opposite effect, by acting as effective cloud condensation nuclei, increasing cloud cover. The relative strength of these effects is hard to predict as the content of black carbon in BBA depends on fuel type and burning conditions, with high black carbon content in the flaming stage and very low in the smoldering stage of fires, e.g. [57]. The mixing state of black carbon in BBA can also vary, e.g. [37] and this can change their radiative effect by a factor of two [31]. To complicate matters further, the microphysical properties of BBA change in time as they interact with other types of aerosols and water vapor [40]. Consequently, understanding the overall effect of BBA on climate needs a characterization of both their global distribution in space and time and their optical and microphysical properties.

Recent development of continental-scale lidar networks provides the opportunity to obtain quality assured optical profiles on a large temporal and spatial scale. For instance, the European Aerosol Research Lidar Network (EARLINET) [52], consists at present of 27 lidar stations which perform coordinated measurements to investigate aerosol properties, transport and modification phenomena. A comprehensive and homogeneous database was created starting on the year 2000, hosting now more than 44.000 profiles [61]. Atmospheric aerosol climatology, Saharan dust outbreaks, forest-fire smoke plumes, photochemical smog and volcano eruptions were intensively studied, e.g. [6–8,39,51]. However the microphysical side of aerosol particles still remains a very extraordinary puzzle for one to put together.

The main difficulty in retrieving microphysical properties arises from the large variability in aerosol composition and physical properties, which cannot be exactly derived from a limited number of optical data. Indeed, our inversion approach involves calculations based on very little piece of information, i.e. average values from optical parameter profiles in specific layers, to construct a whole function of aerosol size distribution. Obviously, tracing back the aerosol size distribution, which resulted our input data (from the lidar), cannot guarantee uniqueness, but this is only part of the situation. Our model relates the optical parameters to the aerosol size distribution through the action of a so-called Fredholm integral operator which is well-known to cause a smoothing effect. This means that the very process of inversion amplifies high frequencies in the input data, i.e. the noisy components become noisier. Adding to this our tenuous knowledge on input data error, there is no wonder for the special care that needs to be taken for a meaningful retrieval.

Regularization methods are a widely preferable choice for an efficient inversion. Algorithms have also been evolving improving the microphysical retrieval process at least for the case of spherical particles, and set at the same time the least required optical parameter setup for the lidar systems. In [41–43], Tikhonov regularization method is used along with the generalized cross validation to handle the contribution of smoothness (penalty term) while in [64] the same regularization method is combined with a modified version of discrepancy principle. Kolgotin and Müller [36] introduce a two dimensional regularization method which makes use of a correlation condition between successive height bins. Böckmann and Kirsche [14] use Padé iteration to regularize the problem and later Osterloh et al. [46] offer an adaptive base point algorithm in addition to the iterative approach. Finally our work is based on a hybrid regularization method firstly presented in [13] and later in [15], which exploits different mathematical features simultaneously and does not require an a priori optical data error assumption.

Artificial data compared to real-life data include no risk of mishandling some of the initial parameters (e.g. the particle radius) as physical entities; dealing with real-life data on the other hand requires their determination. Numerical experiments show that poor selection of these parameters directly affects the solution space quality and compromises the retrieval regardless of the robustness of the regularization algorithm. In this paper we perform sensitivity tests to different data sets, to determine the ideal settings for our algorithm and subsequently produce retrievals in a semi-automated fashion. These parameters involve the refractive index grid and its resolution, upon which the solution space is built, the reasonable radius bounds and mathematical features such as the representation of the solution in a finite dimensional space. The last part of the retrieval procedure is the post-processing taking place right after the regularization is over, involving the appropriate measure (norm) for the fit and the number of solutions for averaging. The initial setup and the post-processing were subject to thorough tests until we confidently apply them to our cases in routine. A real asset to this work was the graphical user interface shown in [49], which allowed us to massively perform these tests minimizing the time factor. The determination of the optimal parameters is followed by the evaluation of our algorithm with the CtoF-AMS and the comparison with AERONET (AErosol RObotic NETwork).

AERONET provides a long-term, continuous database of aerosol optical and microphysical properties extracted by sun photometer measurements and inversions worldwide. The operational protocol of AERONET consists of sky radiances inversions simultaneously at all available wavelengths for the complete solar almucantar or principal plane scenario, referring to radiance at azimuthal angles relative to the sun and at scattering angles away from the sun respectively, together with measurements of aerosol optical depth at the same wavelengths to produce the aerosol size distribution and various microphysical parameters. A direct quantitative comparison of microphysical parameters is often neither possible nor trustworthy, for sun photometer measurements actually consider the entire vertical atmospheric column and do not distinguish specific layers. A detailed description of the inversion advancement and the retrieval products of AERONET is provided in [23–26,60].

Chemical composition and size distribution of submicronic non-refractive ambient aerosols were derived using a CToF-AMS from Aerodyne Research [32]. A careful choice of the measurement cases (Section 2.6) as close to the ground as