Aerosol direct Radiative Effect based on LIdar and Sunphotometer measurements in an Eastern European AERONET/EARLINET Site, ARELISEES

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- **Introduction and motivation**

The role of atmospheric aerosols in Earth's radiative budget is important, but there are still uncertainties in quantifying their radiative effects (IPCC 2007). Aerosols affect the Earth-atmosphere energy budget in different ways: directly, by scattering and absorbing solar and thermal infrared radiation; indirectly, by modifying cloud properties; semi-directly, by heating the atmosphere and evaporating low level clouds. Direct shortwave radiative effect of aerosols is addressed in this project. It depends on aerosol optical properties, in particular aerosol optical depth, single scattering albedo and asymmetry parameter. The uncertainties in modeled aerosol radiative effects are largely due to poor knowledge of their optical properties. Aerosol loading and optical properties are temporally and spatially variable, mainly due to their short lifetimes and spatial variability of their sources and sinks. Their vertical profiles are also variable, and depend on how high they were injected in the atmosphere and on various atmospheric processes. Some studies indicated the importance of knowledge of aerosol vertical profile for estimating their direct radiative effect in cloudy conditions and in the longwave (thermal infrared) spectral range (Yu et al., 2006, and references therein). The importance of this information on modeled shortwave (solar) aerosol radiative forcing in clear sky conditions has also been investigated. Meloni et al. (2005) showed that in the case of strongly absorbing aerosols, aerosol shortwave radiative forcing at the top of the atmosphere (TOA) depends on vertical structure of aerosol layer. Guan et al. (2010) considered weakly and moderately absorbing aerosols and reported that sensitivity of aerosol direct radiative forcing at both the TOA and the surface to a change in vertical profile of aerosol extinction coefficient is not significant. However, they reported that the vertical variability of aerosol extinction coefficient affects vertical profile of the aerosol radiative forcing.

In the frame of the ARELISEES project we conducted lidar, sunphotometer and in-situ measurements at the AERONET/EARLINET site (RADO) in Bucharest, to study variability of aerosol optical properties and to estimate aerosol direct shortwave radiative effects. One of the focuses of the project was also to investigate the sensitivity of modeled aerosol radiative effects on aerosol profile.

- **Scientific objectives**

The objective of this project is characterization of aerosol optical properties and assessment of aerosol shortwave direct radiative forcing and heating rates, in the cases of different airmasses transported to AERONET/EARLINET site in Bucharest. Particular emphasis has been given to investigating the sensitivity of aerosol radiative forcing to vertical profile of aerosol extinction. For that purpose, co-located CIMEL sunphotometer, lidar and aerodynamic particle sizer (APS) have been used as complementary instruments. We focused on the analysis of aerosol optical properties necessary for estimating aerosol radiative effects. Vertically integrated aerosol optical properties (aerosol optical depth, single scattering albedo and asymmetry parameter) have been obtained from the sunphotometer measurements, while lidar measurements were used to derive vertical profile of aerosol extinction coefficient.

- **Reason for choosing station**

  The RADO station has been chosen because it is equipped with a sunphotometer and a multi-wavelength Raman lidar, necessary for our project, as well as in-situ instruments for measuring aerosol properties at the ground level. This site is also interesting because it is affected by aerosols originating from different source regions, including biomass burning from Eastern and Southern Europe and Saharan dust (Nemuc et al., 2011). The period of observation has been chosen due to frequent influence of Saharan dust.

- **Method and experimental set-up**

  We organized an intensive 1-month measurement campaign, from 13 May to 9 June 2012, at RADO (44.348 N, 26.029 E) using a multi-wavelength Raman lidar and sunphotometer. Ground-based size distribution and composition measurements, using APS and AMS, were also conducted to account for Planetary Boundary Layer (PBL) contribution.

  The multiwavelength lidar system (RALI) is based on compact, pulsed Nd:YAG laser, emitting simultaneously pulses of 110, 55 and 65 mJ output energy at 1064, 532 and 355 nm, respectively, with a 10 Hz repetition rate (Nicolae et al., 2010). The receiving Cassegrainian telescope has a primary diameter of 40 cm. A complete overlap between the laser beam and the telescope field of view is expected at the range of 700 m. Photomultiplier tubes (PMTs) are used to detect the received lidar signals in the analog and the photon counting mode, with a corresponding spatial resolution of 3.75m. The detection channels include elastic wavelengths (1064, 532p, 532s, and 355 nm) and Raman wavelengths (607, 387 and 408 nm). Averaging time of the lidar profiles was of the order of 1 min for daytime measurements and 5 min during the night. Optical parameters of aerosols were extracted from lidar data using pre-processing and processing algorithms (Talianu et al., 2007) based on Fernald-Klett method (Klett, 1981; Fernald, 1984) previously tested and validated in the intercomparison campaign of EARLINET-ASOS project (Böckmann et al., 2004).

  CIMEL Sunphotometer is an automatic sun and sky radiometer. The measurement data are centrally processed at GSFC NASA. From direct sun radiance measurements at seven wavelengths (340, 380, 440, 500, 675, 870, and 1020 nm) aerosol optical depth (AOD) is derived, while measurements at 940 nm are used to derive water vapor content. Diffuse sky radiance is measured at four wavelengths (440, 675, 870, and 1020 nm). Aerosol size distributions, refractive indices, single scattering albedo and asymmetry parameter are inferred by inversion of sun and sky radiances.

  Aerodynamic Particle Sizer (APS) was used for ambient air monitoring and measurements of particles from 0.5 to 20 μm in diameter, and Aerosol Mass Spectrometer (AMS) was used for real-time detection of aerosol chemical composition.

  Fu-Liou radiative transfer code (Fu and Liou, 1992) has been used for the calculus of the direct shortwave aerosol radiative effects. It is a four stream broadband model with delta approximation for strong forward scattering of large particles. In this model the solar spectrum is divided into six bands: 200-700 nm, 700-1300 nm, 1300-1900 nm, 1900-2500 nm, 2500-3500 nm, and 3500-4000 nm. Besides aerosols and clouds, the model takes into account the Rayleigh scattering and absorption of O_3, H_2O, O_2, and CO_2. The correlated k-distribution method is used for band-averaging of gaseous absorption coefficients.

- **Preliminary results and conclusions**

  The possibility of synergetic use of the lidar and sunphotometer measurements is subject of ongoing analysis, as it requires testing the sensitivity of the output parameters to various known problems, such as operating time (Raman lidar: night-time, sunphotometer: daytime), lidar overlap, pointing direction, etc. During the measurement period, few profiles with elevated aerosol layers were observed, which are particularly interesting for our project. Although the weather conditions were not favorable most of the time during the measurement period, on several occasions night-time measurements with Raman channels were conducted. In total, 37 one-hour backscatter profiles (both elastic and Raman datasets) were measured during the campaign, and 8 extinction profiles from Raman datasets.

Two interesting cases were identified during the campaign. Lidar range corrected signals for the two selected cases (measurements taken on 22 May and 9 June) are shown in Fig. 1. One-hour profiles of backscatter and extinction coefficients have been calculated.

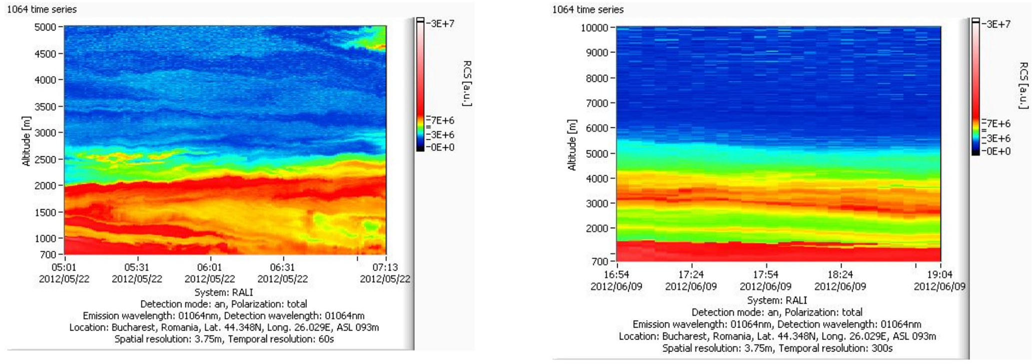

Figure 1. Time series of range corrected signal for the cases of 22 May and 9 June

In both cases AERONET Level 1.5 inversion products show the presence of coarse aerosol mode (Fig. 2), and high single scattering albedo (weakly absorbing aerosols). The lidar-derived vertical profiles of extinction coefficients and corresponding 355-532 nm Ångström exponents shown in Fig. 3, suggest significant decrease of the relative concentration of fine aerosol particles with altitude, up to about 2 km.

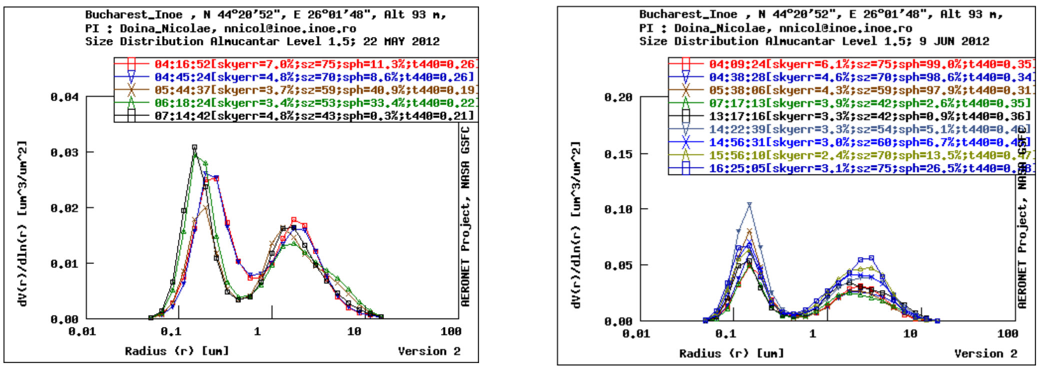

Figure 2. Integrated column size distribution from sunphotometer on 22 May and 9 June

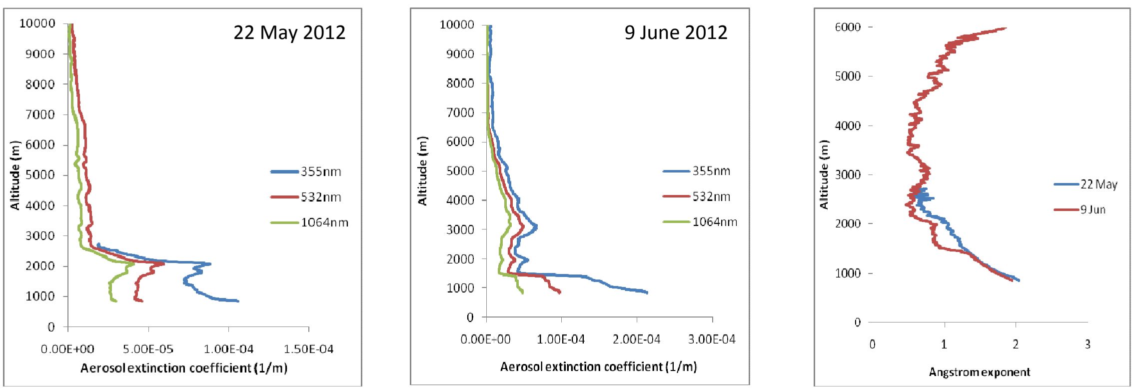

Figure 3. Extinction coefficient and 355-532 nm Ångström exponent profiles for the cases 22 May and 9 June

Lidar (profiles) and sunphotometer (integrated column) retrievals show a possible signature of the mineral dust mixed with continental polluted aerosols, with a pronounced contribution of smoke on 9 June 2012. The presence of dust is confirmed by DREAM model (Fig. 4) for both cases.

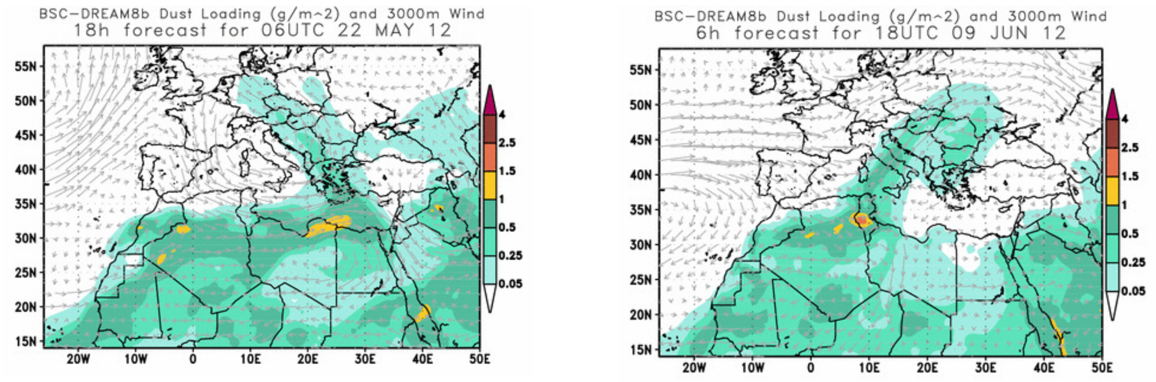

Figure 4. Results of DREAM model on May 22 and June 9

Increased contribution of smoke on 9 June is probably due to the collection of biomass burning aerosols from North Italy, where dense forest fires were present in this period. Increased concentrations of organics and sulfates were sensed on 9 June also at the ground by the C-ToF AMS (Fig. 5), as well as an increased number of fine particles for these species, which are typical for biomass burning.

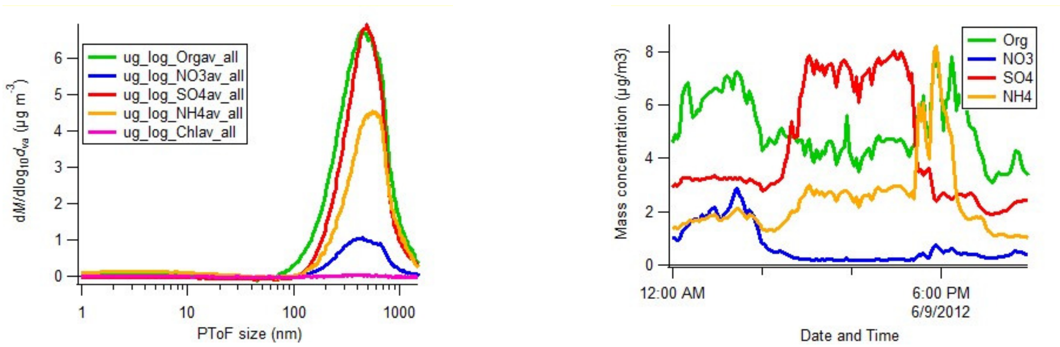

Figure 5. Size distribution and mass concentration of aerosol's components at ground on June 9

Preliminary radiative transfer calculations have been performed for these two cases, using Level 1.5 sunphotometer-derived aerosol optical properties (aerosol optical depth, single scattering albedo and asymmetry parameter). For that purpose they were interpolated/extrapolated at 35 wavelengths between 200 and 4000 nm, after which the spectral averaging within the six solar bands of the code was performed. The vertical profile of aerosol extinction coefficient obtained from the lidar measurements at 532 nm was used, assuming a constant value below the range of the full overlap (below the altitude of 700m). The aerosol direct radiative forcing results are -15.1 and -31.3 W/m^2 at the TOA and -18.28 and -39.5 W/m^2 at the surface, for the cases of May 22 and June 9, respectively. The sensitivity of the aerosol radiative effects to vertical profile of aerosol extinction is subject of current research.

More interesting cases, with two well divided aerosol layers, and influences from Saharan dust, volcanic dust and smoke have also been selected from previous measurements at the site, for study of aerosol radiative effects.

- **Outcome and future studies**

  After obtaining quality assured data, in-depth analysis of aerosol optical properties based on sunphotometer and lidar measurements in combination with APS will be carried out.

  A comprehensive study of aerosol radiative effects based on these measurements, as well as cases selected from previous measurements collected at the site, will be conducted. Particular emphasis will be given to the effect of the assumed vertical profile of aerosol extinction on the modeled heating rates and vertical profile of aerosol radiative forcing.

  It should be noted that using the lidar and sunphotometer measurements in synergy requires test of consistency between them for the cases of interest. This will be done by comparison of lidar-derived aerosol optical depth and Ångström exponent with the corresponding properties obtained from the sunphotometer measurements; cases in which close agreement is reached will be used. Available night-time lidar measurements will be used along with sunphotometer measurements in cases in which temporal and spatial stability of the aerosol layer is observed.

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