# A CASE STUDY OF THE BEHAVIOR OF AEROSOL OPTICAL PROPERTIES UNDER THE INCIDENCE OF A SAHARAN DUST INTRUSION EVENT

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(Received 28<sup>th</sup> Jun 2015; accepted 23<sup>rd</sup> Apr 2016)

**Abstract**. In this paper a local-scale study of Saharan dust loading over Iasi city, Romania, during July 2012 is reported. The intrusion of Saharan dust above the monitoring site [Iasi\_LOASL, Romania (47.19N, 27.55E)] and the variations of ozone and NO<sub>2</sub> concentrations are estimated by using advanced remote sensing techniques (i.e. Aerosol Robotic NETwork – AERONET, Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations – CALIPSO) and forecast models (i.e. DREAM and MAP 3D). The time interval was appropriately selected, to remove the influence of precipitation and clouds on photochemical processes. During intrusion events, the Saharan dust represents the main source for changes in optical and chemical parameters of the atmosphere.

Keywords: remote sensing, AERONET, Saharan dust, aerosols, ozone

### Introduction

Taking into account the major role of aerosols in absorption and scattering of solar radiation, which alters the optical properties of Earth's atmosphere, remote sensing techniques and modeling approach have been recently developed, providing complementary information about temporal and spatial variation of aerosol optical parameters (e.g. Single Scattering Albedo, Ångstrom exponent, Lidar ratio etc.). Recent researches confirmed that in Romania, especially in urban areas, the main sources of air pollution amend the complex mechanisms of photochemical oxidants; they produce an adverse effect on the optical properties of Earth's atmosphere and global climate change (Cazacu et al., 2015, 2011; Papayannis et al., 2014). Of all the classes of photochemical oxidants in the atmosphere, ozone and nitrogen dioxide are among the most extensively

studied; recent studies focused on the possible influence of Saharan dust on them (Ohde and Siegel, 2012).

Actual monitoring and modeling techniques for the Earth's atmosphere (Light Detection And Ranging – LIDAR, sun photometer, UV cameras, MAP3D etc.) provide useful information on changes in the weather conditions and the climate (Belegante et al., 2011; Cazacu et al., 2011; Nicolae et al., 2010; Timofte et al., 2011).

The Saharan dust significantly influences the radiative transfer through absorption, scattering and reflection phenomena, strongly affecting the energy fluxes and spectral distribution of solar radiation (Earth's radiation budget), as well as photosynthesis processes (Haywood et al., 2008; Papayannis et al., 2014). In addition to the influence of Saharan dust, radiative transfer is also affected by anthropogenic sources of nitrogen oxides (NO and NO<sub>2</sub>) and volatile organic compounds (VOCs) under the influence of solar ultraviolet (UV) radiation, favouring photochemical reactions (Ohde and Siegel, 2012; Shon et al., 2008).

This paper presents a study of Saharan dust loading over long distances, as well as monitoring of possible influences on the atmosphere over Romania. The analysis of experimental data and theoretical models specific to Romania have shown fairly low concentrations of Saharan dust, but large enough to influence the optical parameters, so that the transfer of solar radiation is affected even during sunny days.

Some research and development projects, such as ROmanian LIdar NETwork (ROLINET) and Romanian Atmospheric research 3D Observatory (RADO) investigate the air quality in the North-East region of Romania, in the proximity of the Iasi city; several research papers reported on this topic. Starting from the end of spring to early fall, over Romania, multiple Saharan dust events occur (Cazacu et al., 2015, 2012, 2011; Gothard et al., 2014; Labzovskii et al., 2014; Papayannis et al., 2014; Unga et al., 2013). In this paper, we report the influence of Saharan dust during July 2012 on the local air quality. Besides, we compare the air quality forecast given by MAP3D with the experimental data acquired through remote sensing techniques available on the site, the Atmospheric Research 3D Observatory RADO Station of the Alexandru Ioan Cuza University of Iasi (sun photometry). Other experimental and theoretical approaches in the quantification of air quality were also used, i.e. Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), Dust REgional Atmospheric Model (DREAM) and Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT). Evidence of Saharan dust intrusion was provided by sun photometry and by calculus methods of aerosols dispersion (based on databases of various meteorological survey stations).

## Methodology

Mesoscale Air Pollution 3D Modeling (MAP3D) developed at the Laboratory of Environmental Fluid Mechanics and Hydrology of École Polytechnique Fédérale de Lausanne, Switzerland, is capable of providing daily forecasts for air pollutants (O<sub>3</sub>, NO, NO<sub>2</sub>, PM<sub>10</sub> particles) (Couach et al., 2004). The MAP3D is based on the chemical model MM5/CHIMERE, which has been successfully applied for the entire Romanian territory [with a resolution of 15x15 km<sup>2</sup> (Cazacu et al., 2011)].

Complementary researches were based on the following techniques:

- Aerosol Robotic NETwork (AERONET), a NASA network for monitoring and characterizing atmospheric aerosols using a Cimel Automatic Sun Tracking Photometer

CE 318 ground-based, solar- powered, weather-hardy, robotically-pointed sun and sky spectral photometer (Holben et al., 1998). Starting from 7 May 2012, the monitoring station Iasi\_LOASL located in Iasi, Romania (47.19N, 27.55E) became active in this network, giving researchers immediate access to the quantitative values for various types of aerosols (Unga et al., 2013). In order to provide optical aerosol properties the Ångstrom exponent, Single Scattering Albedo (*SSA*), Lidar report calculated by AERONET (*S*<sub>AERONET</sub>) and size distribution were considered from level 1.5 with zenith angle larger than 50°, retrieval error less than 5% and AOT (aerosol optical thickness) at 440 nm greater than 0.2. The Lidar ratio is defined as ratio between extinction coefficient and backscatter coefficient and can be calculated using the SSA parameter (at 532 nm) and phase function at 180 degree (Cattrall et al., 2005).

- Dust REgional Atmospheric Model (DREAM), an operational website developed by Earth Sciences Division of the Barcelona Supercomputing Center, used to confirm the presence of Saharan dust over Romania (Basart et al., 2012; Nickovic et al., 2001; Pérez et al., 2006).

- Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, a complete system for computing both simple air parcel trajectories and complex dispersion, also comprising capabilities for deposition simulations. The model uses the existing meteorological forecast fields from regional or global models to compute advection, stability and subsequent dispersion (Draxler and Rolph, 2015);

- The Cloud Aerosol Lidar with orthogonal Polarization (CALIOP) system, on board of the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) platform, is based on the elastic-backscatter Lidar used for aerosols and cloud investigation. The CALIOP is providing level 1 products, high-resolution profiles of the attenuated backscattered radiation by aerosols and clouds at visible (532 nm) and near-infrared wavelengths (1064 nm) and depolarized backscattered radiation using the visible channel (at 532 nm). With previously developed algorithms (Winker et al., 2009), level 2 products [classification of different features by layer type, e. g. clouds, aerosols type like Saharan or mineral dust, biomass burning, urban/industrial, the extinction coefficient profile and total column aerosol optical depth (AOD) for a defined Lidar ratio for each detected aerosol layer] are estimated. Mamouri et al (Mamouri et al., 2009) and Pappalardo et al (Pappalardo et al., 2010) have shown that level 1 attenuated backscatter profiles are in reasonable agreement with ground-based Lidar measurements.

## **Results and discussions**

The data from July 2012, considered as a hot summer month, were acquired since the aerosols optical properties vary from season to season, as shown by Mihai and Stefan (Mihai and Stefan, 2011). Using data from AERONET and analysing the Ångstrom and SSA parameters, we selected the possible Saharan dust intrusion events. A decrease in the Ångstrom parameter (specially for values lower than 1.5 at 440–870 nm) indicates an increase of number of particles with a diameter greater than 1  $\mu$ m (coarse mode) from the volumetric distributions retrieval while an increase of SSA with increasing wavelength (from 440 nm to 1020 nm, with SSA values between 0.91 and 0.99) indicates a dust influence (Dubovik et al., 2002; Giles et al., 2012). Thus, an analysis of selected days (the 3<sup>rd</sup>, 4<sup>th</sup>, 15<sup>th</sup> and 25<sup>th</sup> of July 2012) presenting this trend was made, and we validated the presence of Saharan dust with data from the HYSPLIT and BSC

DREAM models, as well as data acquired through the AERONET, as will be seen below.

As previously reported by Unga et al. (2013), July 2012 studies on the tropospheric aerosols above Iasi revealed major influence of urban and industrial aerosols on the air quality in this region. Afterwards, Cazacu et al. have proven that the Iasi\_LOASL monitoring site is specific for urban/industrial aerosols type with influences of biomass burning and mineral dust especially when Saharan dust events occur (Cazacu et al., 2015). Thus, in all AERONET data the presence of urban/industrial aerosols was always registered over the studied site.

Concerning the present study, by applying the HYSPLIT model for the days of  $3^{rd}$ ,  $4^{th}$ ,  $15^{th}$  and  $25^{th}$  of July 2012 in which the Ångstrom coefficient had a decreasing behavior below 1.4 (the highlighted dates in *Fig. 1*) and the SSA parameter varied from 0.91 (at 440 nm) up to 0.99 (at 1020 nm) with a very low spherical particle fraction (0.2÷6.6), results indicating the presence of Saharan dust have been obtained (*Fig. 2*).



Figure 1. Variation of Ångstrom parameter during July 2012.

It can be observed that these intrusion events are originating from Sahara desert. These results are in agreement with the BSC DREAM model, confirming the presence of Saharan dust for  $15^{\text{th}}$  and  $25^{\text{th}}$  of July 2012 (*Fig. 3*). Even though for the days of  $3^{\text{rd}}$  and  $4^{\text{th}}$  of July 2012 the BSC DREAM model does not agree with the presence of Saharan dust above the observation site, particle trajectories indicated by HYSPLIT suggest the Sahara desert as possible origin of measured. The presence of Saharan dust for these days was previously shown by Unga et al (2013) taking into account the Lidar ratio from AERONET data. The given uncertainties are ruled out by comparison with

previous observation quantities such as: the Ångstrom coefficient, the Single Scattering Albedo (*SSA*), and the LIDAR report calculated by AERONET.



Figure 2. Air mass trajectories from HYSPLIT model.

All the above-mentioned parameters indicate that in fact we are dealing with Saharan dust, since significant drops of the Ångstrom coefficient from 1.5 to 0.5 and  $S_{AERONET}$  from 50 sr to 30 sr are recorded. Although the concentration is rather small, the optical quantities measured by the solar photometer correspond to those reported for Saharan

dust class-type dust (Ansmann et al., 2011, 2009; Muller et al., 2010, 2009; Unga et al., 2013).

By analyzing data gathered for high altitudes, the local influence can be disregarded, thus obtaining additional information on the influence of Saharan dust over the local air quality. The concentration is rather different and quantitative analysis provided by the BSC DREAM indicates variations of the surface density from 0.05 to 0.50 g/m<sup>2</sup> over the observation site.

As can be seen from *Figure 3*, 15 and 25 July 2012 as clear days and Saharan dust influence days are confirmed by AERONET (*Fig. 1*) and HYSPLIT (*Fig. 2*). The absence of cloudiness is confirmed by both the BSC DREAM model and the meteorological data using the EUMETSAT satellite database - European Organization for the Exploitation of Meteorological Satellites (EUMETSAT, 2012). The above-mentioned models and experimental data are required to use the MAP3D chemical model.



Figure 3. Saharan dust loading by DREAM forecast for July 15<sup>th</sup> and July 25<sup>th</sup>, 2012.

As it can be shown for the  $3^{rd}$ ,  $4^{th}$ ,  $15^{th}$  and  $25^{th}$  July 2012, the MAP3D model forecasts an increase of the PM<sub>10</sub> concentration (*Fig. 4*). These results are calculated for troposphere, indicating the daily maximum values for altitudes up to 8 km. The complete series from July 2012, indicates that PM<sub>10</sub> concentration increased over the entire tropospheric column (up to 8 km).

Besides, as it can be easily observed, a new concentration increase is forecasted starting with 19 July. By appealing to HYSPLIT and BSC DREAM models, we cannot confirm any additional intrusion of Saharan dust. The data acquired on 19 July indicate the strong necessity of gathering data from as many sources as possible.

Since the MAP3D model indicated an increase in PM10 concentration for 24 to 25 July and the DREAM model forecasted a powder density similar to that recorded on the 15 July (*Fig. 3*), these data were completed with products derived from the CALIPSO Lidar measurements. The total backscattering coefficient recorded by CALIPSO for the 00:35 to 00:48 interval (UTC) is given (*Fig. 5 - up*). The marked area represents the Lidar signal from atmospheric constituents. Appealing to inversion methods for calculation of optical parameters using the Lidar signal, a classification of the aerosols is given (*Fig. 5 - down*). From the data gathered by CALIPSO, a dispersion of the Saharan dust up to 5 km altitude can be emphasized. One can state that acquired experimental data match the forecast data.



*Figure 4.* Variation of daily maximum concentration of PM<sub>10</sub> for July 2012 from MAP3D model.



*Figure 5.* (up): 532 nm backscatter Lidar profile from CALIPSO and (down): aerosol type classification.

The influence of the Saharan dust storms over the local climate can be quantified at last. Recent studies indicate variations of ozone concentrations during the Saharan dust events (Frouin and McPherson, 2012; Ohde and Siegel, 2012). A decrease in the ozone concentration was explained by the fact that Saharan dust absorbs in the wavelength range below 440 nm. Another observation indicates a decrease in UV solar radiation intensity reaching the ground level, while the infrared spectrum is not affected. Based on these observations, a parameter closely monitored was the tropospheric ozone concentration.

In our case, the influence of Saharan dust over the tropospheric ozone production was not observed. Given that the data were gathered during clear days, the influence of atmospheric clouds can be excluded. As can be inferred from *Figure 6*, the variation of the tropospheric ozone concentration may be explained by the following equations (Cazacu et al., 2011; Seinfeld and Pandis, 2006):

$$[NO_{3}+h\nu(\lambda < 700nm)] \rightarrow NO + O_{2}$$
$$[NO_{3}+h\nu(\lambda < 7580nm)] \rightarrow NO_{2} + O$$
$$[NO_{2}+h\nu(\lambda < 420nm)] + O_{2} \rightarrow NO + O_{3}$$

As instance, the 3D distribution of the ozone given by the MAP3D model is not uniform up to altitudes as high as 8 km, as a consequence of the typical chemical reaction of ozone in the atmosphere (*Fig.* 7). Furthermore, no observable influence of the Saharan dust was noticed over our ground observatory, due to its small concentration.



Figure 6. Variation of  $NO_2$  and  $O_3$  concentrations during July 2012 forecasted by MAP3D model (ground level).



*Figure 7. Three-dimensional distribution of*  $O_3$  *computed by* MAP3D *model (up to 8 km).* 

### Conclusions

The impact of Saharan dust in low and middle troposphere on the behavior of aerosol optical properties was emphasized using modern remote sensing techniques (sun photometer, CALIPSO) and modelling approaches (MAP3D, HYSPLIT, DREAM models).

On the  $3^{rd}$ ,  $4^{th}$ ,  $15^{th}$  and  $25^{th}$  of July 2012, AERONET data show values between 0.6 – 1.5 of Ångstrom parameter and values between 0.91  $\div$  0.99 of SSA at an increase of wavelengths from 440 nm to 1020 nm. Although these values indicate the presence of mineral dust, the AERONET Lidar ratio (e.g. 30 sr), HYSPLIT data, DREAM model and CALIPSO satellite data confirmed also the influence of Saharan dust load over Iasi\_LOASL monitoring site.

The influence of Saharan dust on the tropospheric ozone production has not been clearly observed, probably because of strong absorption of UV-vis radiation ( $\lambda$ <420 nm) by dust particles. The variations of ozone and nitrogen dioxide concentrations are not directly related to Saharan dust loadings and only some seasonal variations are emphasized.

Further complementary measurements are required to fully understand the impact of Saharan dust on the ozone molecules at different altitudes in the troposphere using UVmolecular emission spectroscopy. Acknowledgements. This work was supported by the Romanian Space Agency (ROSA) within Space Technology and Advanced Research (STAR) Program (Project nr. 98/29.11.2013) and by the strategic grant POSDRU/159/1.5/S/137750, Project "Doctoral and Postdoctoral programs support for increased competitiveness in Exact Sciences research" cofinanced by the European Social Found within the Sectorial Operational Program Human Resources Development 2007 – 2013.

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