CHEMICAL ENGINEERING TRANSACTIONS, Volume 16, 2008
Guest Editor Eliseo Ranzi
ISBN 978-88-95608-03-7 ISSN 1974-9791

Analysis of aerosol’s long-range transport using synergetic techniques

C. Radu, D. Nicolae, A. Nemuc, C. Talianu
National Institute of R&D for Optoelectronics INOE 2000,
1 Atomistilor, P.O. Box MG-5, Magurele, Bucharest, Romania

Aerosol particles of the Earth’s atmosphere participate in the global energy balance directly by scattering and to a lesser extent by absorbing incoming solar radiation. They also have an influence on the climate indirectly as they act as cloud condensation nuclei onto which cloud droplets are formed (Houghton *et al.* 2001). The regional nature of aerosols makes tropospheric aerosol trends more difficult to determine when long-range transport is involved.

In this paper, several studies based on lidar and complementary data are presented. Qualitative and quantitative analysis of local aerosols – characteristics and distribution – and also of aerosols transported in the free troposphere is described. The synergy between lidar, sunphotometer and numerical models output have proven to be a suitable tool to distinguish Saharan dust from smoke and anthropogenic pollution.

Introduction

Earth radiation budget is strongly influenced by atmospheric aerosols. The global energy balance is influenced by aerosol particles directly by scattering, and much less by incoming solar radiation absorption. Recent studies on the impact of aerosols by direct and indirect effects on the radiative forcing in a global average show that they are of the same order of magnitude as the warming effect (Forster, P., et al., 2007). To quantify the radiative effect of aerosol on climate, global and regional climate models are used. Haywood M. et.al (1999) found out that, without taking into account the aerosols, the energy budgets from a global climate model does not match observations from the Earth Radiation Budget Experiment (ERBE). There are studies that include prediction of aerosols online in numerical weather prediction (NWP) models (Pérez C. et al. (2006)). But, there still are uncertainties regarding the indirect and direct effects, which are related with the aerosol influence on climate.

Formation of new atmospheric aerosol particles (Kulmala M. et al. (2003)) has been observed worldwide, but the whole mechanism of this formation is still under debates. Ground based measurements of aerosol properties - chemical composition, scattering and absorption, size distribution - are performed in several locations, many of them in Western Europe, and only few in Eastern Europe. These in situ measurements are used also for validation of global models, by considering aerosol concentrations at the surface or by providing significant information about local trends, chemical composition and variability on various time scales. Unfortunately, in situ ground based measurements have the disadvantage of small number of monitoring sites over the Europe and limited number of the measured parameters due to their position near the surface. Since there are differences in meteorological conditions and because in situ measurements are dependent of conditions mostly at or near the surface while the direct and indirect radiative forcing depend on the aerosol vertical profile, a comparison

Please cite this article as: Radu C., Nicolae D., Nemuc A. and Talianu C., (2008), Analysis of aerosol’s long-range transport using synergetic techniques, Chemical Engineering Transactions, 16, 345-352

between in situ measurements and global atmospheric models measurements are complicated. For example, the spatial resolution of global model grid boxes is a few degrees of latitude and longitude and the time steps for the atmospheric dynamics and radiation calculations may be minutes to hours depending on the process to be studied. This introduces limitations at comparison with observations performed over smaller spatial extent and shorter time duration.

Lidar measurements are a suitable solution to this problem, since they provide optical (and in some cases also microphysical) properties of particles with high spatial and temporal resolution (Ansmann A. et al. (1992)) as vertical profiles. Due to the complicated operation and cost of the equipment, lidars are only sparse in the territory and by consequence can provide information only at local level. Actual trend is to elaborate a schedule over a long period of time for the spatially relevant networks of state-of-the art instruments, in order to collect data to be further used for statistical analysis and model optimization and validation. A step forward is represented by networks of measurement instruments like AERONET (for the surface based remote sensing sun photometer (Holben, B. N. (2001) et al.) or EARLINET (for the aerosol Lidar systems (Bösenberg, J. et al.(2003))).

Methodology

Lidar (Light Detection And Ranging) is an active remote sensing technique based on the emission of laser pulses (ns) into the atmosphere and the analysis of the return signal. Depending on the emitted and selected wavelength at the detection, different characteristics of air pollutants can be measured. The photons scattered by the atmospheric particles are collected by a receiving optical telescope. To ensure a good wavelength selection of the lidar signals, narrow-band interference filters are used, one for each detected wavelength (1064 and 532 nm). The selected radiation is focused on a photomultiplier (PMT) – for visible and UV light, respectively on an avalanche photodiode (APD) – for IR light, which convert the optical signal into electrical signal. The distance between the lidar system and the atmospheric target can be calculated measuring the delay time between the emitted and the received laser pulses. Thus, range resolved measurements of the desired air pollutants or atmospheric parameters can be obtained.

The Lidar system used for measurements has a maximum range of 8-10 km and a 3 m range resolution. The emitted output energies per pulse are 50 and 100 mJ (for 532 and 1064 nm), with a 20 Hz repetition rate. The backscattering signals are collected through an optical telescope, and further acquired and digitized in the analog and/or the photon counting mode using fast transient recorders and then transferred to a computer for storage and analysis. Because Nd:YAG lasers have prove their efficiency from energetic and pointing stability point of view, most of aerosol lidar systems are built starting from the fundamental and the 2^{nd} and/or 3^{rd} harmonics of the Nd:YAG laser.

Sun photometer measures the radiance at seven wavelengths using almucantar and principle plane scenarios. The almucantar scenario measures radiance at azimuthal angles relative to the sun, but sky radiances in the almucantar are not sensitive to aerosol vertical variations in the case of single-scattering approximation. On the other way, the principle plane scenario measures radiance at scattering angles away from the sun. The combination between radiance data and aerosol optical depth measurements and estimations of land and water surface reflectance can be inverted to estimate aerosol optical properties. The signal can be inverted to provide aerosol optical properties such as the complex index of refraction, single scattering albedo, size distribution, and phase functions (Holben et al., 2001).

This paper presents a synergy between lidar measurements, sun photometer data and atmospheric modeling in order to determine the type and origin of aerosols which travel over the Romanian territory having an significant impact on the regional radiative budget. Lidar technique was used to determine range-resolved vertical profiles of aerosols optical parameters (such as the backscatter and extinction coefficient) with very high spatial and temporal resolution. The altitude of layers as well as their temporal evolutions were observed and further used as inputs in air mass trajectories models. By a proper choice of input parameters (layer altitude, time interval), the identification of aerosol source was possible. In several cases (two of which are shown here), the origin of aerosol was found to be at significant distance from the destination. These particles have a different structure and composition than local aerosols, but this cannot be measured directly by an elastic backscatter lidar. On the other hand, detection of light at multiple wavelengths can provide – by a suitable inversion technique – some information about the microphysical properties of such particles, which can be used to make a proper classification.

The AERONET type sun photometer (7 wavelengths) was used to make an assessment of aerosol type for these special cases. Due to the multiple wavelengths detection and the inversion technique, the size distribution, complex refractive index and Angstrom exponent of aerosols - averaged in the air column – can be derived. All these parameters are an indicator of the aerosol type and, implicitly, of their probable origin. By combining the information obtained from lidar (layer altitudes), forecast models (probable temporal interval of transport from the source to the destination), transport models (air mass trajectories at layer altitudes and in the temporal interval) and microphysical properties derived from sun photometer measurements, a proper classification of aerosols (type and origin) can be done.

In some particular cases, due to the mixing of aerosols coming from different sources, the identification of both type and origin is not possible. This is due to the fact that data acquired with the sun photometer represent in fact the average return of entire air column, insensitive to layers of different provenience. In this situation, the derived microphysical properties are not typical for a specific aerosol, thus conclusions cannot be drawn.

Our study also underlines that not always the results obtained by all methods are consistent. In general, forecast models are much less accurate and must be used carefully. Experimental data must be the first choice. As we mentioned before, models are an important tool in such analysis, but their outputs are trustable only if they are coincident with measurements. In any case of long range transport, before taking any decision on aerosol origin, the precipitation field along air mass path must be checked, in order to exclude the possibility of wet deposition.

Results

Two cases are presented in this paper: one refers to a smoke intrusions coming from large forest fires in the vicinity of Romanian border, and the other to a Saharan dust event, both measured over the Magurele-Bucharest site.

Range Corrected Signal (RCS) plots (Fig. 1a and b) of the lidar signal shows in both cases layers between 2000 and 3500 meters of altitude, over the PBL (Planetary Boundary Layer). The distinction between PBL and free troposphere layers is clear for both cases.

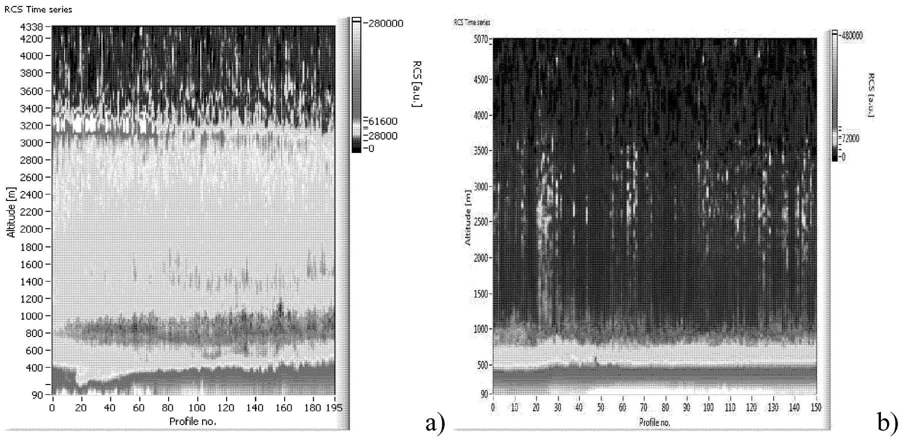


Figure 1. The RCS over Bucharest a) on 20 July, b) on 30 August

The altitude of layers is even more evident in the backscatter coefficient profiles (Fig. 2a and b) obtained by the inversion of the 30 min averaged lidar signal. The graphs bellow show the high backscatter coming from the high concentration of aerosols in the PBL but also a significant backscatter in the free troposphere around 3000m. For the lower part of the profiles, it is assumed that only local aerosols contributes, but the presence of particles in the free troposphere, visible in the upper part of the profiles, must be further investigated.

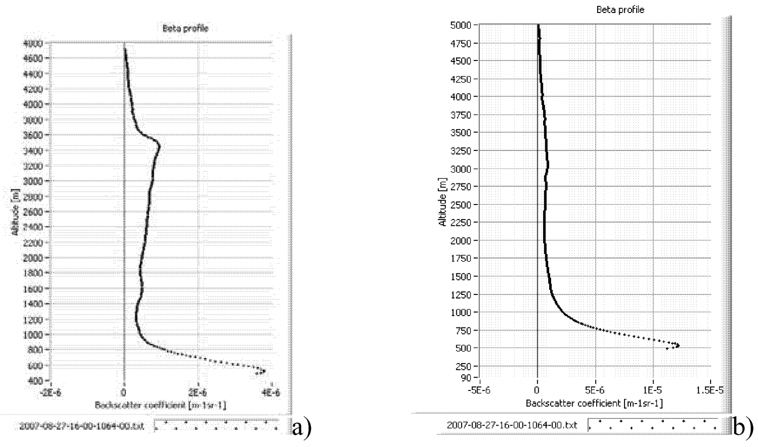
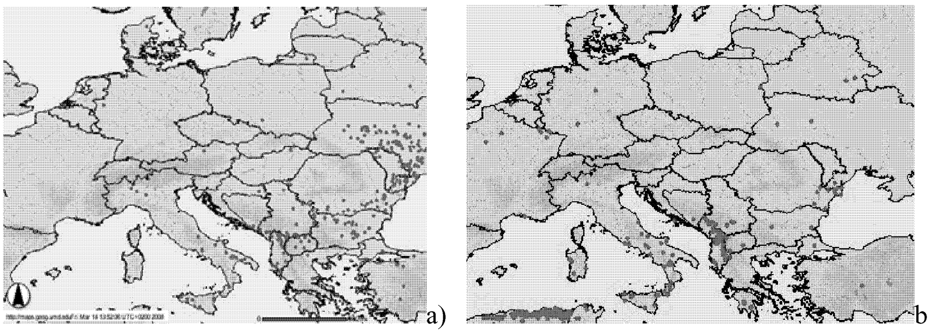


Figure 2. The backscatter coefficient at 1064 nm in Bucharest on: a) 20 July and b) 30 August

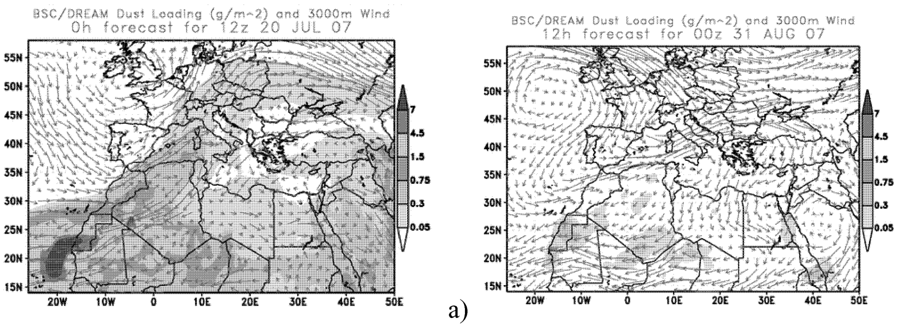
First step in the assessment of aerosol origin is to investigate the 2 major sources which are known to have contributions in the area: forest fires and Sahara outbreaks. In Fig. 3, MODIS maps for the 2 cases are shown. It can be seen that fire spots are present in both cases, much more dense in the first case although. Depending on the air mass trajectories, contributions of these fires to the aerosol layers measured by lidar can be suspected, but not confirmed.



a) b)

Figure 3. MODIS fires on: a) 20 July and b) 30 August

On the other hand, DREAM (Dust REgional Atmospheric Model.) forecast for the days in this study (20 July and 30 August) show no probable Saharan dust intrusion in southern part of Romania (Fig. 4a and b). Using only these 2 data sources, it appears that the aerosol layers measured by lidar can only be smoke particles, coming from nearby fires. The type and source of these layers cannot be obtained from elastic backscatter lidar data, so other confirmation are necessary. In case of major pollution sources identification, the advantage of the air mass back-trajectories tracing method is that covering of very large areas by the investigating system is not necessary. If the measurements are regular at local level and if some meteorological parameters are known (speed and direction of winds, temperature and humidity), with a good probability can be evidenced aerosol sources at large distances from the measurement point, if their contribution is important. Using the altitudes of the layers as inputs in HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) and 6-days backward trajectories, probable origin or aerosols can be identified (Fig. 5a and b).



a) b)

Figure 4. DREAM prognosis model for a) 20 July, b) 30 August

It can be seen from Fig. 5a that in the first case (20 July), the air masses arriving at 2000m and 3000m altitude over Bucharest have the same path for 3-4 days. They both cross Albania, Moldavia and Ukraine, regions were important forest fires took place, conform with MODIS maps. This event started on 17 July up to 21 July. For the first 2 days, air masses coming from Ukraine and Moldavia reached the Romanian territory, and traveled back to Ukraine. After 19 July, the circulation of air masses changed, traveling from Ukraine to Romania and further to Greece. During the entire period, none of the paths is coming from Sahara region. Consequently, the most probable aerosol type for this case is smoke aerosol.

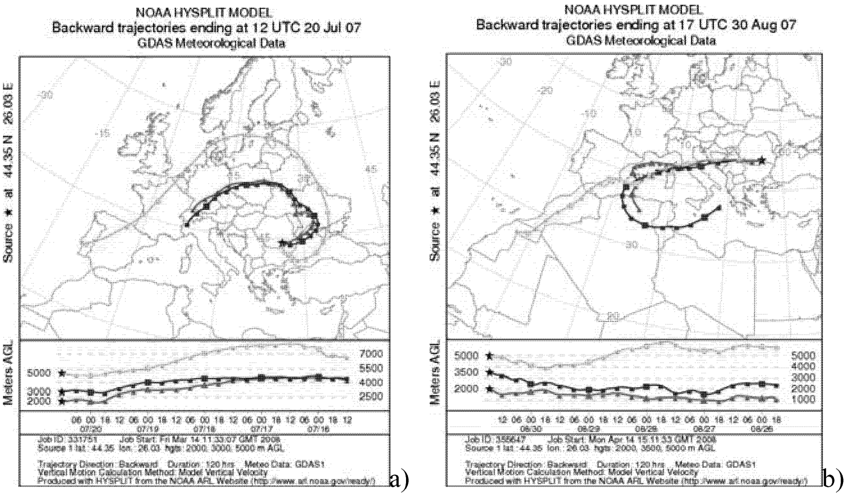


Figure 5. HYSPLIT back-trajectories for the selected cases:a) 20 July 12.00 UTC and b) 30 August, 17.00 UTC

Due to the fact that extracting microphysical information from lidar data is not always possible and moreover this is not a warranty that a proper classification of aerosol can be done, another important confirmation of aerosols type and origin was made based on data collected from sun photometers co-located with lidar.

Conclusion regarding the smoke-type composition of aerosol layer above PBL on 20 July is confirmed by the sun photometer derived data, which shows a bi-modal size distribution, having both modes of the same magnitude (Fig. 6 a). It must be noted that Bucharest local aerosol has a quite different aspect for the size distribution (Fig. 6 b). The derived complex refractive index for the air column averaged aerosol for the first case is about $n = 1.45 + 0.15 \cdot i$. The important value of the imaginary part of the refractive index indicates a high proportion of soot, which is again consistent with the assumption made on the aerosol origin.

Another parameter which can be used to distinguish between different types of aerosols is the asymmetry factor. Saharan dust, for example is essentially a non-spherical particle, so that its asymmetry factor will have higher values than the corresponding value for the smoke particles. In our case, the retrieved asymmetry factor for 20 July is about 0.55. This value excludes the possibility of dust or marine type aerosol, but is still a question of choosing between urban or smoke type aerosol.

By combining all information for this case, it can be concluded that a smoke layer coming from Moldavia and Ukraine has reached Bucharest atmosphere on 20 July. This layer is visible in lidar measurements above PBL and influences sun photometer data, although its contribution in the average column is probably not very important (large concentrations of urban aerosols in the PBL have a much higher impact).

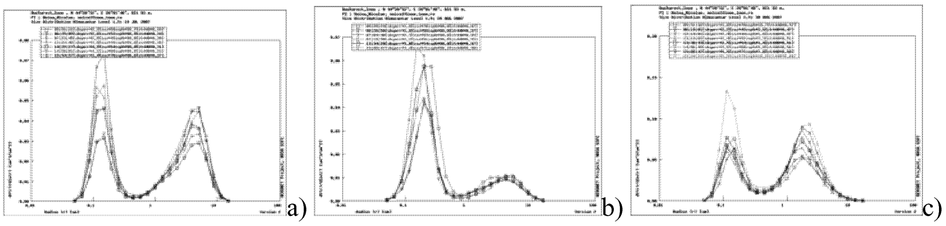


Figure 6 Size distribution of air column average aerosol
a) 20 July – smoke; b) 28 August – urban low absorption; c) 30 August - dust

As we mentioned before, in the second investigated case lidar measurements also show aerosol layers above PBL (Fig. 1b and 2b). From MODIS and DREAM images (Fig. 3b and 4b), it appears that it can only be smoke, since fires are present around Romania but no Saharan dust outbreak is forecasted. Air mass trajectories at 2000m and 3000m (Fig. 5 b), on the other hand, did not cross fire areas (concentrate in the South Europe), but the Central part of Europe, where no important fire is visible. Looking even backward, it can be seen that trajectories at all 3 selected altitudes have the same origin: North Africa. HYSPLIT graph shows a continental influence bellow 2000 m altitude and possible influences from Albanian fires at higher altitudes. On 30 August, air mass trajectories starting from Sahara travel over the Algerian fires al lower altitudes (below 2000 meters) and further on over the Mediterranean Sea, Italy and Serbia, adding some continental influences. At this point, a new assumption rises, that aerosols over PBL could have dust structure, even if DREAM model didn't forecast any intrusion in our part of Europe.

To investigate this case, sunphotometer data are again of great help. In Fig. 6c, the aerosol size distribution derived from the inversion of 7-wavelengths detection is shown. One can see that the aspect of this function has no significant differences from the one of smoke. The same bi-modal characteristic, of approximately equal height lobs, is present in both cases. The imaginary part of the complex refractive index, although, has a much lower value than in the case of smoke: $n = 1.45 + 0.006 \cdot i$. This means that the proportion of low absorption components in this type of aerosol is much higher than in case of smoke, or even urban, and the proportion of soot is reduced. Saharan dust, which is mineral, satisfies this condition. It is also in good agreement with the origin of air masses, indicated by HYSPLIT. A supplementary confirmation comes from the value of the asymmetry factor, which is around 0.65 for the case on 30 August. This is an indicator of the aerosol shape and in this case shows important asphericity, again suitable for mineral dust.

Conclusions

Lidar is a powerful tool for the study of aerosol long-range transport. Main advantage of lidar is the real time observation of aerosol layering, which can be further used to identify the origin and the path of air mass. This technique has its limitations, but in combination with modeling and complementary techniques more data can be obtained. Using lidar and sun photometer in combination with satellite imagery, forecast and transport models we were able to make an assessment of aerosol type and properties, as well as to localize the sources of long-range transport aerosols which affected air quality over the Bucharest area. Good agreement between various techniques was achieved. The synergy of all these techniques proved to be a suitable tool for the retrieval of information about aerosol origin, type and distribution.

Aknowledgements

DREAM aerosol dust data were provided by the Earth Sciences Division, Barcelona Supercomputing Center, Spain. Part of this work was also financed by the EARLINET-ASOS European Union Project under contract RICA-025991. Air mass back trajectories were produced with the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT-4.6) model (NOAA). AERONET data were provided by NASA. We thank Philippe Goloub for data analysis of sun photometer data from AERONET site INOE-Magurele).

References

[1]. Forster, P., et al., 2007: Changes in Atmospheric Constituents and in Radiative Forcing. *In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change* [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor and H. L. Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, USA.

[2]. Haywood M., Ramaswamy, V., and Soden, B. J.: *Tropospheric aerosol climate forcing in clear-sky satellite observations over the oceans*, *Science*, 283, 1299–1305, 1999.

[3]. Pérez C., S. Nickovic, G. Pejanovic, J. M. Baldasano, and E. Ozsoy 2006, *Interactive dust-radiation modeling: A step to improve weather forecasts*, *J. Geophys. Res.*, 111, (D16), D16206, doi:10.1029/2005JD006717, 2006.

[4]. Ansmann A., et al., 1992: Combined Raman Elastic-Backscatter lidar for vertical profiling of moisture, aerosol extinction, backscatter, and lidar ratio, *Applied Physics* **B55**, pp. 18-28.

[5]. Kulmala M., Vehkamaki, H., Petaja T., dal Maso M., Lauri A., Kerminen V.-M., Birmili W. and McMurry P.H., *Formation and growth rates of ultrafine atmospheric particles: A review of observations*, *J. Aerosol Sci.*, 2003

[6]. Holben, B. N., et al., *An emerging ground-based aerosol climatology: Aerosol optical depth from AERONET*, *J. Geophys. Res., 106*, 12,067–12,098, 2001.

[7]. Bösenberg, J., et al., *A European aerosol research lidar network to establish an aerosol climatology*, *MPI-Report 317*, Max-Planck Inst. für Meteorology, Hamburg, Germany, 2003.

[8]. Draxler, R.R., and G. D. Hess, *An overview of the Hysplit_4 modeling system for trajectories, dispersion, and deposition*, *Aust. Meteorol. Mag.*, 47, 295– 308, 1998.

[9]. http://maps.geog.umd.edu/activefire_html/checkboxes/eur_checkbox.htm