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Air mass modification processes over the Balkans area detected by aerosol Lidar techniques

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Anthropogenic aerosols may have a substantial effect on the present-day aerosol abundance, optical depth and thus, may play an important role on the forcing of climate. This study combines aerosol lidar measurements with atmospheric modeling in order to assess the origin and type of aerosols affecting air quality in the Balkans region, and thus, having an impact on a possible modification of regional radiative budget. Based on the climatological aerosol data set containing the spatial and temporal evolution of the vertical distribution of aerosols over Greece and Romania, derived from lidar measurements, an analysis of two case studies is presented: one evidencing smoke aerosols traveling over the Balkan area, and the second evidencing transport of Saharan dust particles.

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1. Introduction

The impact of aerosol particles on the Earth's climate due to their effect on the radiation budget is of strong interest and the subject of many recent studies.[1][2][3] Aerosol particles in the Earth's atmosphere participate in the global energy balance directly by scattering, and to a lesser extent absorb incoming solar radiation. They also have an influence on the climate indirectly as they act as a cloud condensation nuclei (CCN) onto which cloud droplets are formed [4]. The radiative effect of aerosol on climate has been quantified in global and regional climate models. For example, [5] Haywood (1999) found that energy budgets from a global climate model would not mach observations from the Earth Radiation Budget Experiment (ERBE), unless aerosols were taken into account. Fewer studies have included prediction of aerosols online in numerical weather prediction (NWP) models [6]. The uneven distribution of aerosols on a regional scale makes tropospheric aerosol trends more difficult to determine than trends in long-lived trace gases. Moreover, there are too few long-term records of tropospheric aerosols, mostly in the free troposphere.

On the other hand, the aerosol particle population is in a constant turmoil. The particles are colliding with each other, gaseous compounds are condensing onto them altering their composition, chemical reactions are occurring on the surface and within the particles, particles are lost due to deposition to the various surfaces in the Earth-atmosphere interface, and the population is renewed as new particles are formed either directly or via diverse gas-to-particle conversion mechanisms. Formation of new atmospheric aerosol particles has been observed worldwide [7], but the exact mechanism by which this formation occurs is scientifically still an open question. Surface-based measurements of aerosol properties such as size distribution, chemical composition, scattering and absorption are performed at a number of sites, either at long-term monitoring sites, or specifically as part of intensive field campaigns, mostly in Western and Central Europe, but very rarely over Eastern Europe[8][9]. These *in situ* measurements provide essential validation for global models, for example, by constraining aerosol concentrations at the surface and by providing high quality information about chemical composition and local trends. In addition, they provide key information about variability on various time scales. Drawbacks are related with the number of such monitoring sites, but also with the restricted number of parameters that can be measured, generally only near ground. Comparisons of *in situ* measurements against those from global atmospheric models are complicated by differences in meteorological conditions and because *in situ* measurements are representative of conditions mostly at or near the surface while the direct and indirect radiative forcing depend on the aerosol vertical profile. For example, the spatial resolution of global model grid boxes is typically 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 poses limitations when comparing with observations conducted over smaller spatial extent and shorter time duration. A possible solution is to establish spatially relevant networks of state-of-the art instruments following a strict measurements schedule over a long period of time, in order to collect data which can be afterwards used for statistical analysis and model optimization and validation. From this point of view, a significant advancement since

GAW (Global Aerosol Watch Program) [10] is the continued deployment and development of surface based remote sensing sun-photometer sites such as AERONET (Aerosol Robotic Network) [11], and the establishment of networks of aerosol lidar systems such as the European Aerosol Research Lidar Network (EARLINET) [12], the Asian Dust Network (ADNET) [13], and the Micro-Pulse Lidar Network (MPLNET) [14].

2. Methodology

Lidar (Light Detection And Ranging) technique is an active remote sensing method based on the emission of short laser pulses (ns or fs duration) to the atmosphere under study and the analysis of the return signal [15]. According to the air pollutant or the atmospheric parameter to be studied, different wavelength(s) have to be selected, both for the transmission and reception. The backscattered laser photons by the atmospheric volume under study are collected by a receiving optical telescope. The wavelength selection of the lidar signals is performed by a set of spectral narrow-band interference filters or a high resolution spectrometer. Photomultipliers (PMTs) and/or avalanche photodiodes (APDs) are used to detect the backscattered photons at the respective wavelengths (from the ultraviolet to the near-infrared). Measuring the delay time between the emitted and the received laser pulses one is able to calculate the distance of the probed atmospheric volume and thus perform range resolved measurements of the desired air pollutants or atmospheric parameters. Lidar signals are acquired and digitized in the analog and/or the photon counting mode by fast transient recorders and subsequently transferred to a personal computer for further analysis and storing. In the case of atmospheric aerosols measurements, wavelengths ranging from the ultraviolet to the infrared region (0.355 to 12 μm) can be selected, depending on the application. Due to the proved energetic and pointing stability of Nd:YAG lasers, most aerosol lidar systems are built starting from the fundamental and the 2nd and/or 3rd harmonics of the Nd:YAG laser.

Sun photometer radiance measurements can be inverted to produce aerosol optical properties such as size distribution, single scattering albedo, phase functions, and the complex index of refraction. Sun photometers measure the radiance at four or more wavelengths using almucantar and principle plane scenarios. The almucantar scenario measures radiance at azimuthal angles relative to the sun. For at least single-scattering approximation, sky radiances in the almucantar are not sensitive to aerosol vertical variations. The principle plane scenario measures radiance at scattering angles away from the sun. [11] These radiance data in combination with aerosol optical depth measurements and estimations of land and water surface reflectance are inverted to estimate aerosol optical properties.

This study combines lidar measurements with atmospheric modeling in order to assess the origin and type of aerosols which travels over Balkan region, having an impact on modification of the regional radiative budget. The capability of the lidar technique to derive range-resolved vertical profiles of aerosols optical parameters (backscatter and extinction coefficient) with very high spatial (10-15 m) and temporal resolution (a few seconds up to a few minutes) was used to identify the altitude of layers and the temporal evolution of intrusions. Using these altitudes as inputs in atmospheric models, the source of aerosols was identified. Several confirmations from additional techniques (sun photometry, satellite remote sensing, forecast or transport models) were also obtained for selected case-studies.

Two lidar systems were used in this study, one located in Athens, Greece (at NTUA) and one in Bucharest, Romania (INOE). NTUA's lidar system is a multiwavelength lidar operating at 1064, 532 and 355 nm and detects both elastic backscatter signals (at 1064, 532 and 355 nm) and Nitrogen Raman signals (at 607 and 387 nm)[16] INOE,s lidar system is a 2-wavelength elastic backscatter lidar operating at 1064 and 532nm. [17] Data processing algorithms used for data inversion at both stations were previously tested and validated in the intercomparison campaign of EARLINET-ASOS project [18]. Lidar combined with meteorological data were used to quantify the long-range transport and deposition mechanisms involved and to identify the major aerosol sources affecting air quality over the Balkan region. Synchronized and regular (routine) aerosol lidar observations were performed in order to give a novel insight into the air mass modification, to deduce the position and the contribution of major aerosol sources in Balkan region, and to predict future trends over the studied area. This was possible by using the aerosol back-trajectory tracing method, HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) [19]. It is, also, a method to distinguish between local originated dust – which is transported on medium distances by troposphere winds - and Saharan dust transported on long distances by stratospheric winds.

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. By using data provided by the two systems situated at a considerable distance one to each other, the Romanian and Greek teams were able to cover the entire area between them, which means an important part of the Balkans region.

3. Results and discussion

Two case studies are presented in this paper, one evidencing smoke aerosols traveling over the Balkans area, and the second evidencing transport of Saharan dust particles. In order to assess the origin of aerosols, we used MODIS [20] data for major fires and HYSPLIT back-trajectories. HYSPLIT forward-trajectories were also used in order to follow air masses traveling to destination. 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 lidars. In this way, three essential different methods are used to validate the assumption of aerosol's source and class: lidar and sun photometer measurements and atmospheric modeling.

Case 1-Forest fires aerosols: 17-24 July 2007

During this period, a large number of forest fires in Ukraine and Moldavia were visible from MODIS data (Fig. 1). Instead, no Saharan dust intrusion in Balkans was forecasted by the Dust REgional Atmospheric Model (DREAM) [21] (Fig. 2). Lidar measurements over Bucharest show distinct aerosol layers around 3000 m for the entire period (Figs. 3 and 6), but these layers became visible over Athens only after 20 July (Fig. 6). In the figures bellow, a comparison between the backscatter coefficient at 1064nm measured in Bucharest (gray line) and Athens (black line) is presented, for the two abovementioned periods. The presence of aerosol layers in the free troposphere is even more visible in the color coded representation of Square Range Corrected Signal (RCS), which evidence not only the altitude of the layer, but its temporal evolution too (Figs. 7 to 10).

Fig. 1. Fires in Ukraine and Moldavia, 17-18 July 2007.

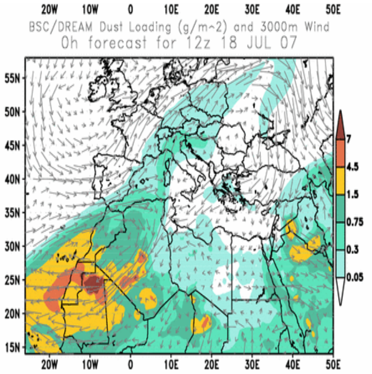

Fig. 2. Saharan dust distribution in Romania and Greece, 17-18 July 2007.

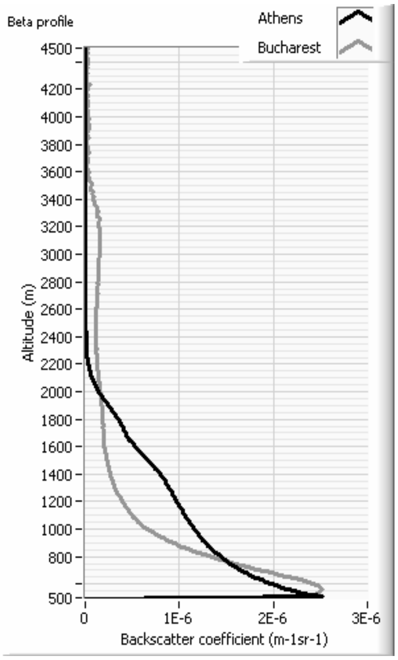

Fig. 3. Backscatter coefficient at 1064 nm in Athens and Bucharest (17 July 2007).

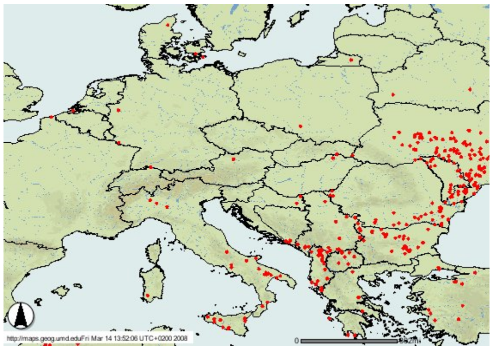

Fig. 4. Fires in Ukraine and Moldavia, 20-21 July 2007.

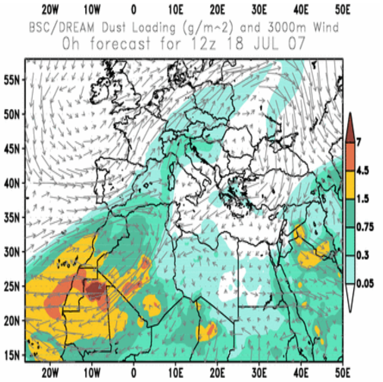

Fig. 5. Saharan dust distribution in Romania and Greece, 20-21 July 2007.

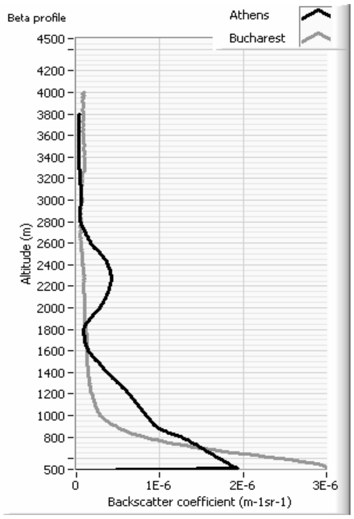

Fig. 6. Backscatter coefficient at 1064 nm in Athens (21 July 2007) and Bucharest (20 July 2007).

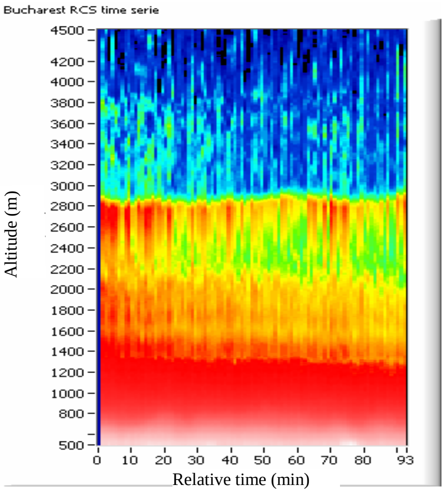

Fig. 7. RCS over Bucharest, 17 July 2007, starting 17:00 UT.

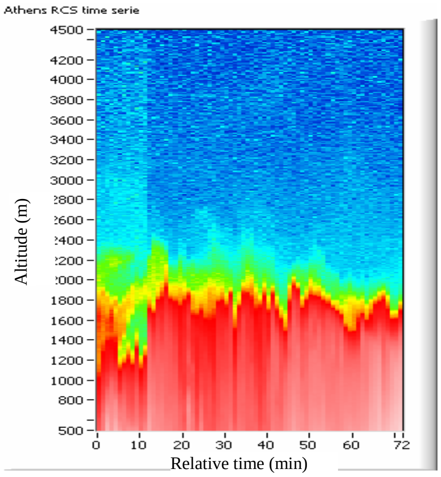

Fig. 8. RCS over Athens, 17 July 2007, starting 17:00 UT.

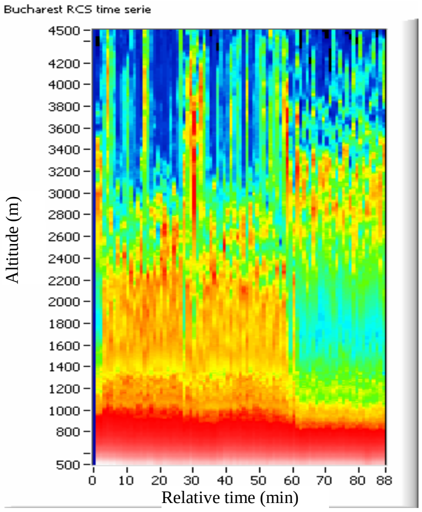

Fig. 9. RCS over Bucharest, 19 July 2007, starting 12:00 UT.

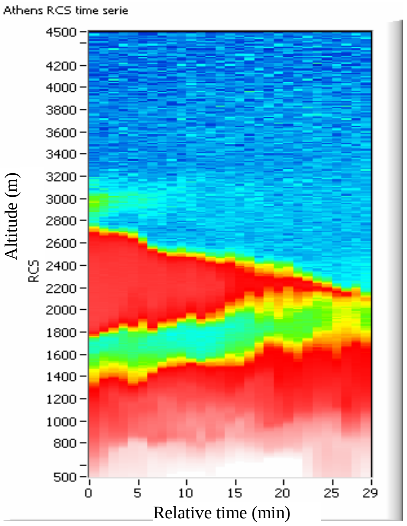

Fig. 10. RCS over Athens, 21 July 2007, starting 12:00 UT.

During this event, air masses coming from Ukraine and Moldavia reached the Romanian territory, and traveled back to Ukraine for several days. After 19 July, the circulation of air masses changed, traveling from Ukraine to Romania and further to Greece. This situation is confirmed by the HYSPLIT code. To run the program, specific altitudes of aerosol layers evidenced by lidar, were used. In the Fig. 11 it can be seen that air masses arriving over Romania on 17 July at 3000 m altitude, crossed an area characterized by a dense distribution of fires (Ukraine and Moldavia), while air masses arriving at higher altitudes are coming from Hungary where no fires were present. Fig. 12 demonstrates that in the following 2 days the same air masses went back to Ukraine. This result is consistent with lidar measurements over Bucharest, which showed a persistent aerosol layer above PBL (Fig. 3), but also with lidar measurements in Athens, which show no intrusion of aerosols in the free troposphere (Fig. 3). A completely different situation begun on 19 July, when air masses coming from Ukraine and traveling over Romania changed their trajectory towards Bulgaria and Greece. The HYSPLIT code shows the same path of air masses over Ukraine and Romania, collecting smoke aerosols (Fig. 13), but this time the 3000m altitude air masses went to Bulgaria and Greece (Fig. 14). This result is again consistent with our lidar measurements, both over Bucharest (Fig. 6) and Athens (Fig 6). This time, both lidars detected aerosols in the free troposphere, dispersed in a larger altitude interval in the case of Bucharest, and concentrated around 2500 m altitude in case of Athens. A possible explanation of the disperse aspect of aerosols over Bucharest could be the influence of air masses arriving at 2000 m and 5000 m also from regions where important fires (North of Romania) were present. The confirmation of aerosol type comes from Sun photometer data operating in Bucharest, which classified the integrated column of aerosol as smoke high absorption during the entire period [22].

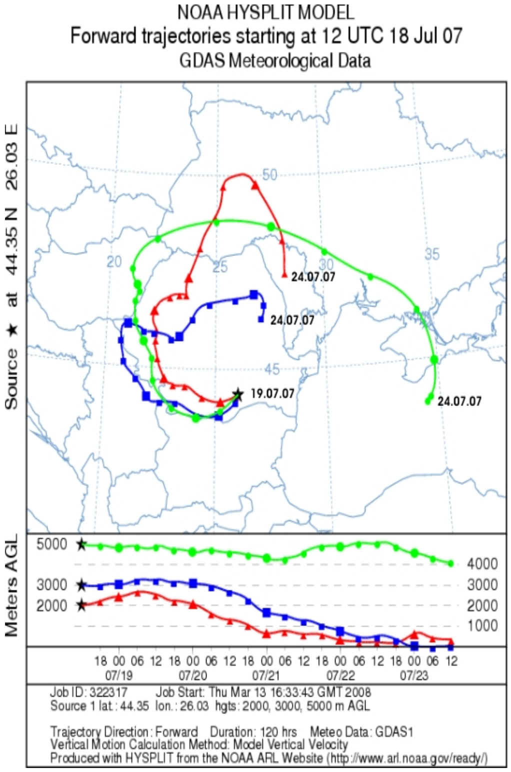

Fig. 12. Air mass forward- trajectories starting from Romania on 18 July 2007.

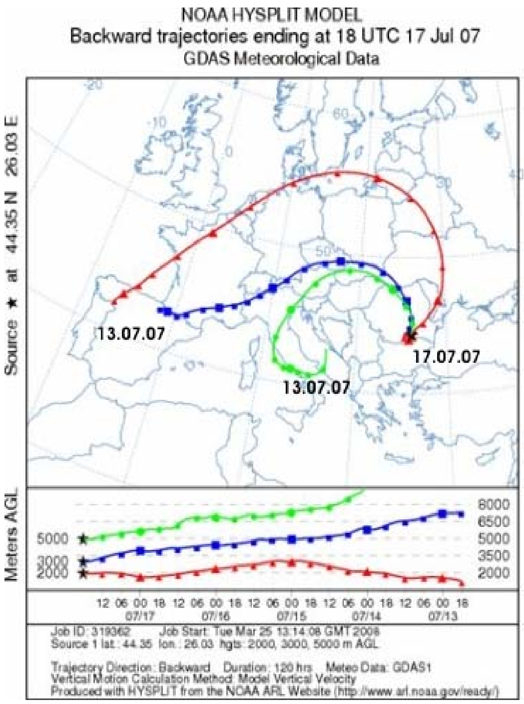

Fig. 11. Air mass back-trajectories ending over Romania on 17 July 2007.

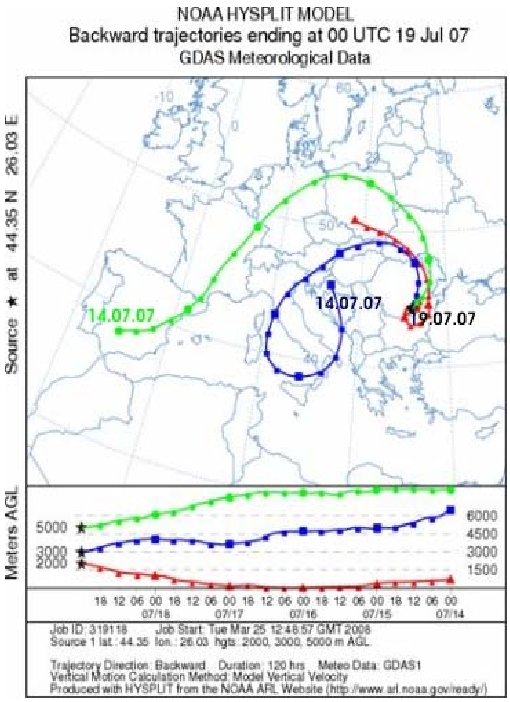

Fig. 13. Air mass back-trajectories ending over Romania on 19 July 2007.

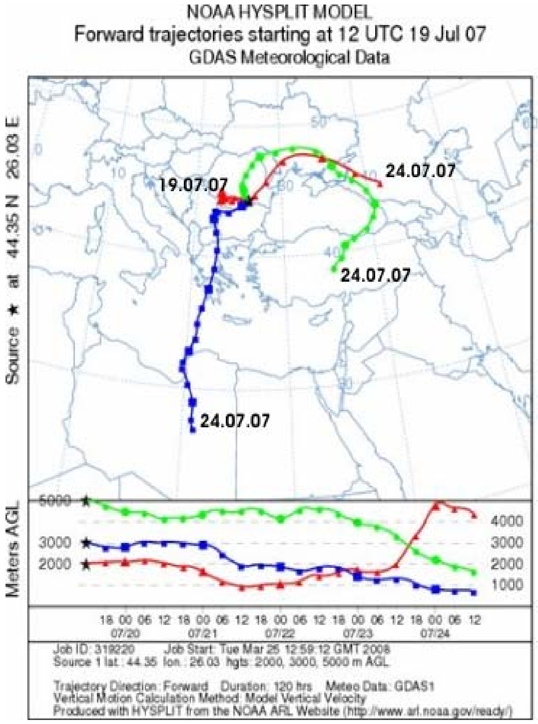

Fig. 14. Air mass forward-trajectories from Romania, on 19 July 2007.

Case 2-Desert dust aerosols: 25-30 June 2007

Another aspect of aerosol climatology in Balkans is related to the intrusion of Saharan dust particles. This kind of events occurs more often in Greece then Romania, due to the typical paths of air masses coming from Africa, crossing Spain, Italy and Mediterranean Sea 23. Nevertheless, during spring and summer periods, the massive injection of the Saharan dust into the troposphere combined with the lack of precipitation increases the probability for this type of aerosol to reach Central and Eastern Europe, including Romania. Such an episode was recorded during 25-30 June 2007 at the location of both lidar stations: Bucharest and Athens. In the figures bellow it can be seen directly from the RCS lidar time series (Figs. 15, 16), but also from the backscatter coefficient profiles (Fig. 17), the direct presence of an important aerosol layer around 2500 m altitude (over Bucharest), respectively of 2 aerosol layers at 2000 m and 3000 m height (over Athens). Aerosol layers were persistent between 25-30 June over Greece, but were removed by wet deposition on 28 June in Romania. This event was successfully forecast by DREAM model (Fig. 19). No important fires on air masses paths were evidenced by MODIS data (Fig. 18).

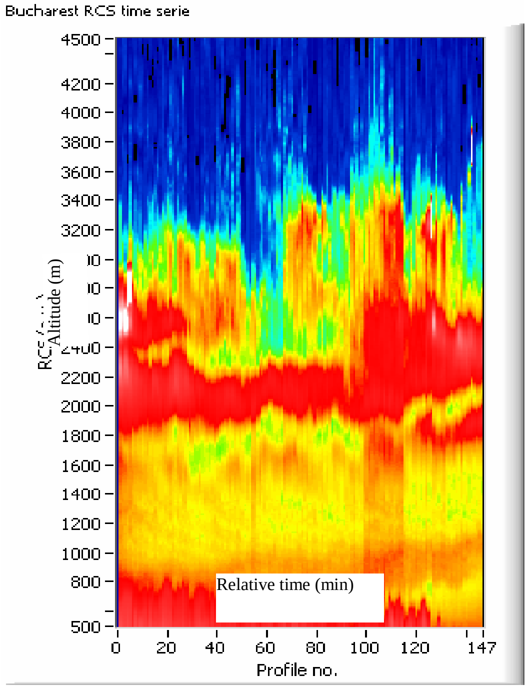

Fig. 15. RCS over Bucharest, 28 June 2007, starting 00:00 UT.

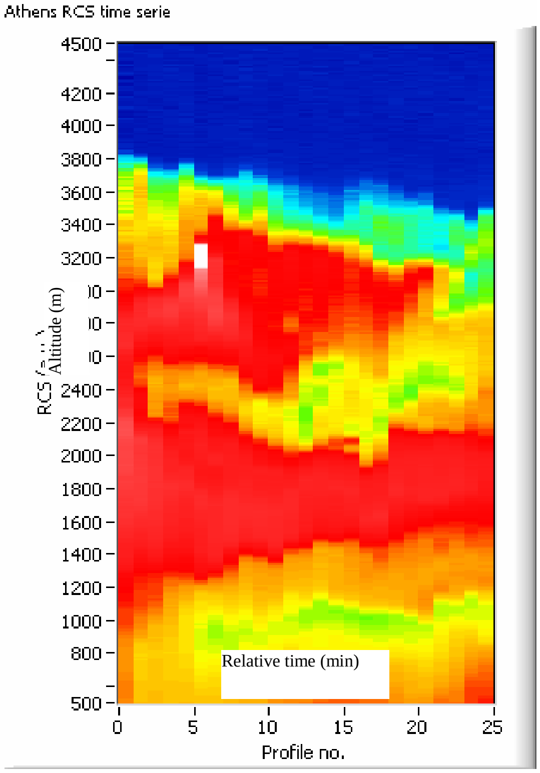

Fig. 16. RCS over Athens, 29 June 2007, starting 03:00 UT.

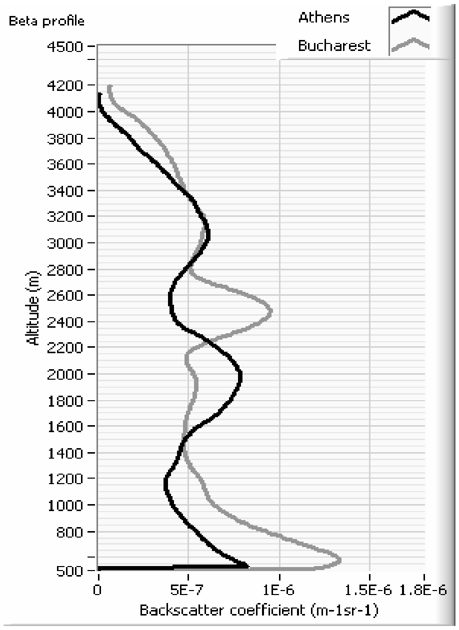

Fig. 17. Backscatter coefficient at 1064 nm over Athens (29 June 2007) and Bucharest (28 June 2007).

It must be noted that, even having the same source (Sahara desert), air masses reaching Romania have different paths then air masses reaching Greece. As shown in Fig. 20, air masses which entered Romania followed approximately a straight line, traveling over South Europe (Spain, Mediterranean Sea, Italy, Serbia) and being influenced, in consequence, by continental pollution too. Nevertheless, the 7-wavelength inversion of sun photometer data evidenced values for the microphysical parameters of integrated column aerosol typical for dust particles.

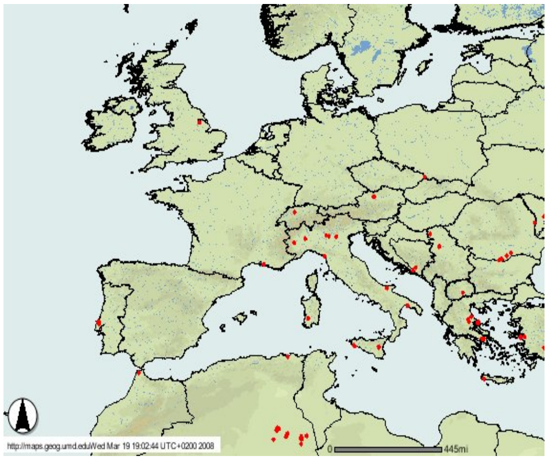

Fig. 18. Fires in Greece and along the Danube river (25-28 June 2007).

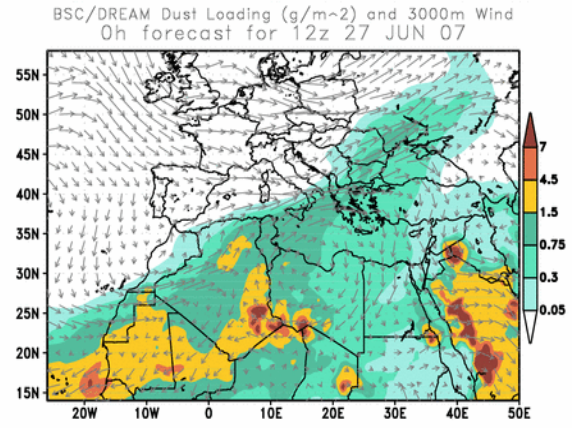

Fig. 19. Dust distribution over Romania and Greece (27 June 2007).

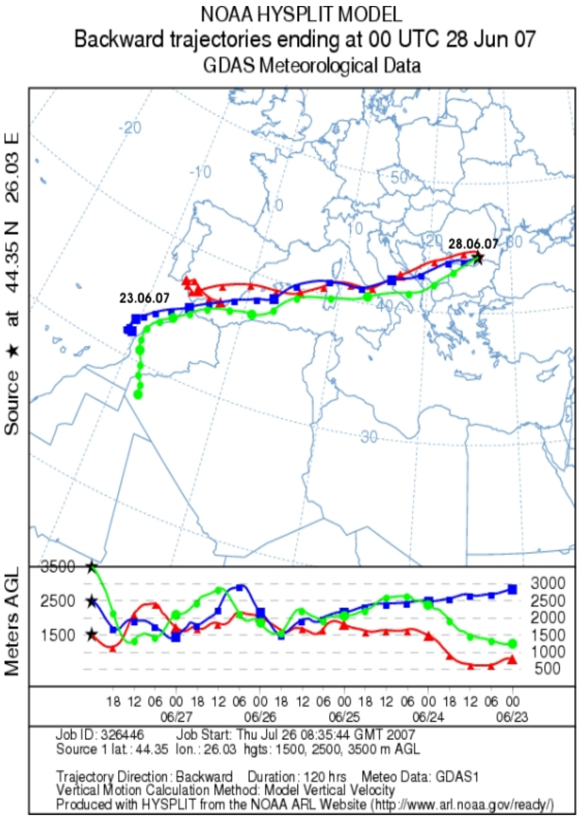

Fig. 20. Air mass back-trajectories ending over Romania, on 28 June 2007.

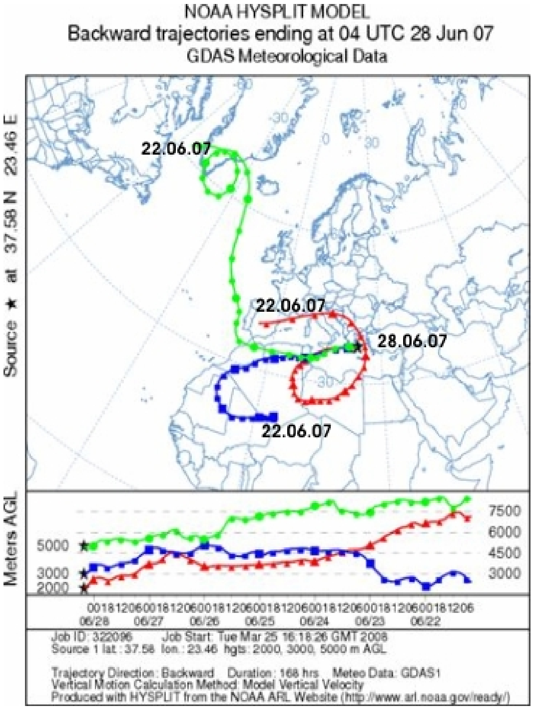

Fig. 21. Air mass back-trajectories ending over Athens on 28 June 2007.

In case of aerosols reaching Athens, the HYSPLIT code showed trajectories that crossed North Africa (Fig. 21). As confirmed by lidar, two distinct layers at 2000m and 3000m were attributed to aerosols coming from the Saharan region.

4. Conclusions

The lidar technique was proved to be a suitable 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 such as sun photometer, important information about aerosol origin, type and distribution can be derived. In this paper we presented a two-case analysis of aerosol transport processes affecting air quality data over the Balkans region: one evidencing transport of forest fires aerosols and a second evidencing transport of Saharan dust particles over Romania and Greece. These data came from a central climatological aerosol data set on the spatial and temporal evolution of the vertical distribution of aerosols over Greece and Romania using laser remote sensing (lidar) technique. The lidar data were analyzed in combination with meteorological and modeling data. Air mass back-trajectory analysis by HYSPLIT code combined with coordinated aerosol lidar profiles obtained over Athens and Bucharest were used to analyze in detail and localize the main sources for aerosols over Europe and Africa affecting air quality over the Balkans regions, and more specifically in Athens and Bucharest.

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References

[1] M. J. Kay, M. A. Box, J. Geophys. Res. **105** (12), 221 (2000).

[2] J. Hansen, L. Nazarenko, R. Ruedy, M. Sato, J. Willis, A. Del Genio, D. Koch, A. Lacis, K. Lo, S. Menon, T. Novakov, J. Perlwitz, G. Russell, G. A. Schmidt, N. Tausnev, Science, **308**, 1431 (2005).

[3] Intergovernmental Panel on Climate Change (2007). Media Advisory: IPCC adopts major assessment of climate change science. Accessed March 29, 2007.

[4] P. Forster, et al., 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, H. L. Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

[5] M. Haywood, V. Ramaswamy, B. J. Soden, Science, **283**, 1299 (1999).

[6] C. Pérez, S. Nickovic, G. Pejanovic, J. M. Baldasano, E. Ozsoy 2006, J. Geophys. Res., **111**, (D16), D16206, doi:10.1029/2005JD006717, 2006.

[7] M. Kulmala, H. Vehkamaki, T. Petaja, M. dal Maso, A. Lauri, V. -M. Kerminen, W. Birmili, P. H. McMurry, J. Aerosol Sci., 2003.

[8] Performance of DREAM dust model during the

SAMUM (Saharan Mineral Dust Experiment), 2006 Field Campaign, K. Haustein, C. Pérez, J. M. Baldasano, D. Müller, M. Tesche, A. Schladitz, V. Freudenthaler, B. Heese, M. Esselborn, B. Weinzierl, K. Kandler, W. v. Hoyningen-Huene, E. Bierwirth

[9] ACCENT_AIDA - Atmospheric Composition Change: The European Network of Excellence - AIDA aerosol chamber facility, 2006 Field Campaign, Universities Facility for Atmospheric Measurement (UFAM)

[10] www.nilu.no/projects/CCC/tfmm/dessau/galion_dessau.pdf

[11] B. N Holben, D. Tanre, A. Smirnov, T. F Eck, I. Slutsker, J. Geophys. Res., **106**, 12 067 (2001).

[12] J. Bösenberg, V. Matthias, A. Amodeo, V. Amoiridis, A. Ansmann, J. M. Baldasano, I. Balin, D. Balis, C. Böckmann, A. Boselli, G. Carlsson, A. Chaikovsky, G. Chourdakis, A. Comeron, F. De Tomasi, R. Eixmann, V. Freudenthaler, H. Giehl, I. Grigorov, A. Hågård, M. Iarlori, A. Kirsche, G. Kolarov, L. Komguem, S. Kreipl, W. Kumpf, G. Larchevêque, H. Linné, R. Matthey, I. Mattis, A. Mekler, I. Mironova, V. Mitev, L. Mona, D. Müller, S. Music, S. Nickovic, M. Pandolfi, A. Papayannis, G. Pappalardo, J. Pelon, C. Pérez, R. M. Perrone, R. Persson, D. P. Resendes, V. Rizi, F. Rocadenbosch, J. A. Rodrigues, L. Sauvage, L. Schneidenbach, R. Schumacher, V. Shcherbakov, V. Simeonov, P. Sobolewski, N. Spinelli, I. Stachlewska, D. Stoyanov, T. Trickl, G. Tsaknakis, G. Vaughan, U. Wandinger, X. Wang, M.Wiegner, M. Zavrtanik, C. Zerefos, 2003, MPI-Report **348**, Max-Plank-Institut für Meteorologie, Hamburg.

[13] T. Murayama, N. Sugimoto, I. Uno, K. Kinoshita, K. Aoki, N. Hagiwara, Z. Liu, I.Matsui, T. Sakai, T. Shibata, K. Arao, B.-J. Sohn, J.-G. Won, S.-C. Yoon, T. Li, J. Zhou, H. Hu, M. Abo, K. Iokibe, R. Koga, Y. Iwasaka, J. Geophys. Res., **106** (D16), 18,345 (2001).

[14] E. J. Welton, J. R. Campbell, J. D. Spinhirne, V. S. Scott, Proc. Lidar Remote Sensing for Industry and Environmental Monitoring **4153**, Sendai, Japan, SPIE, 151 (2001).

[15] R. M Measures, Laser Remote Sensing. Fundamentals and Applications, Krieger Publishing Company, Malabar, Florida, 1992

[16] GSRT **-** Greek - Romanian Cooperation, National Technical University of Athens. Project Title: “Climatology of aerosols and air mass modification processes in the Balkan area (Athens & Buchurest) using the LIDAR technique “ (2006-2008).

[17] D. Nicolae, C. P. Cristescu, J. Optoelectron. Adv. Mater., **8** (5), 1781 (2006).

[18] Böckmann C., U. Wandinger, A. Ansmann, J. Bösenberg, V. Amiridis, A. Boselli, A. Delaval, F. De Tomasi, M. Frioud, A. Hågård, M. Horvat, M. Iarlori, L. Komguem, S. Kreipl, G. Larchevêque, V. Matthias, A. Papayannis, G. Pappalardo, F. Rocadembosch, J.A. Rodriguez, J. Schneider, V. Shcherbakov, M. Wiegner, Applied Optics **43** (4), 977 (2004).

[19] R. R. Draxler, G. D. Hess, Aust. Meteorol. Mag. **47**, 295 (1998).

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

[21] S. Nickovic, G. Kallos, A. Papadopoulos, O. Kakaliagou, J. Geophys.l Res. **106** (18), 113 (2001).

[22] H. D. Kambezidisa, D. G. Kaskaoutis, Atmospheric Environment **42**, 1892 (2008).

[23] A. Papayannis, D. Balis, V. Amiridis, G. Choudakis, G. Tsaknakis, C. Zerefos, A. Castanho, S. Nickovic, S. Kazadzis, J. Grabowski, Chem. Phys. **5**, 2065 (2005).

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