

LIDAR INVESTIGATIONS OF THE TROPOSPHERE PERFORMED DURING SUMMER 2011 MEASUREMENT CAMPAIGN

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Abstract. Lidar investigations of the troposphere, made over the city of Sofia from May to August 2011, are described. Regular EARLINET weekly measurements were performed in that period as well as everyday lidar observations of the atmosphere during the eruption of the Grimsvötn Island volcano and during Saharan dust incursion. Lidar measurements are performed with an aerosol lidar equipped with Nd:YAG laser. Lidar data are presented in terms of vertical atmospheric backscatter coefficient profiles and color maps of the aerosol stratification time evolution. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) backward trajectories and DREAM (Dust REgional Atmospheric Model) forecasts are employed to make conclusions about atmospheric aerosol's origin. It was concluded that the atmosphere above Sofia was not polluted with volcanic aerosols ejected during the Grimsvötn volcano eruptions from 21-28 May 2011. However, thick urban aerosol layers over Sofia have been registered during the summer months. Reported experimental examples are extracted from regular lidar investigations of the atmosphere within the frame of European project EARLINET.

Key words: lidar, aerosols, clouds, Saharan dust, troposphere.

Introduction

Aerosols, very small particles in the air, play an important role in the global climate changes and influence on the ecological state of the environment. They are a significant source of direct and indirect effects on the planetary radiative budget. Atmospheric particles determine the positive or negative radiative forcing in function of its size, chemical composition, spatial and temporal distribution. The height and size distributions of aerosols are critical to all climate influences. Atmospheric particles have a direct radiative forcing because they scatter and absorb solar and infrared radiation in the

atmosphere. They also alter the formation and precipitation efficiency of liquid-water, ice and mixed-phase clouds, thereby causing an indirect radiative forcing associated with these changes in cloud properties. Aerosol-cloud interaction is recognized as one of the key factors influencing cloud properties and precipitation regimes across local, regional, and global scales and remains one of the largest uncertainties in understanding and projecting future changes (J.E. Penner et al., 2001; Guibert, S. et al., 2005; Solomon, S. et al., 2007).

There is a natural aerosol component consisting mostly of soil dust, sea salt, biogenic and organic matter that is geographically and seasonally variable. Among the different types of aerosols, Saharan dust is the dominant natural components in the troposphere (Gian Paolo Gobbi et al., 2000; Perez L. et al., 2008, Sassen K. et al., 2003, U. Pöschl, 2005). Similarly, major volcanic eruptions, which occur infrequently and presumably randomly, inject large quantities of volcano ash and gases into the atmosphere at significant heights. Winds can transport the ash and gases rapidly and in multiple directions which depend on the wind speed and wind direction so that transport is possible over long distance in just a few hours (Ansman A. et al., 1997; Hansell A. L. et al., 2006; A. Schreiner et al., 2004; Kelly P. M. et al., 1996). There is also an anthropogenic component that is linked to fossil-fuel and biomass burning, as well as other human activity. This component has been steadily increasing with global industrialization and urbanization. In highly populated and industrialized regions and in areas of intense agricultural burning the aerosol forcing is much stronger than the globally averaged one, contributing to the global warming. Aerosols, when concentrated near the surface, have long been recognized as affecting pulmonary function and other aspects of human health (N. Sabbagh-Kupelwieser et al., 2010; E. Katragkou et al., 2009; Tasić M. et al., 2006). Urban air consists of a significant fraction of sub micrometer and ultra fine particles (urban aerosols, smog), which give a small contribution to the particulate mass, but are said to be associated with a number of significant negative influences on human health. Smog is a type of air pollution derived from vehicular emission, internal combustion engines and industrial fumes. It can be formed in almost any climate where industries or cities release large amounts of air pollutions. During periods of sunny weather, when the upper air is warm enough to inhibit vertical circulation in the atmosphere, the smog resides for a longtime near the ground, over densely populated cities or urban areas and can reach dangerous levels. Such event is often observed in geologic basins encircled by hills or mountains.

Aerosol effects on the environment are very hard to quantify due to the fact that the amount and origin of atmospheric particles vary substantially with location and from year to years, and in many cases exhibit strong seasonal variations. Extending the principle of radar to the optical range, lidar (**L**ight **D**etection **A**nd **R**anging) technology has found use in many of today's aerosol investigations. The fast spread of lidars contributes to their organizing in lidar networks as EARLINET (European Aerosol Research Lidar Network) (EARLINET:<http://www.earlinet.org>). At present, 27 lidar stations distributed over Europe are part of the network. Main result of such cooperation is the establishment of a quantitative lidar dataset describing the aerosol vertical, horizontal, and temporal distribution, including its variability on a continental scale. Additional more specific measurements (on Saharan dust, volcanic ash, forest fire) are also included in the project work program (Papayannis A. et al., 2008). The joint analysis of lidar observations in

different locations is a useful approach for better understanding and interpretation of some regional aerosol events and their influence on the environmental conditions. Bulgarian lidar station at Sofia (Laser Radar Laboratory of the Institute of electronics, BAS) has been involved in coordinate regular measurements according to the schedule of the EARLINET project. Under appropriate weather conditions, we carry out measurements twice a week. Two night time measurements are performed every Monday and Thursday after sunset, while daytime measurements are performed every Monday at noon when the Sun is in its zenith. These times are selected in order to have one daytime measurement at a well-developed atmospheric planetary boundary layer (PBL) condition, and two observations with low-background light to monitor the aerosol layer developed during the day. When special aerosol loading is forecast, the regular schedule of measurements is altered and more observations are conducted in view of obtaining as full as possible an image of the vertical distribution and temporal changes of the aerosol field over lidar station. Usually each lidar measurement lasts for 1-3 hours. The lidar signal is accumulated for 5-10 min (corresponding to data accumulation of 600-1200 different raw profiles received at each laser pulse). These profiles are subsequently summarized to cover 30-min time of observation in conformity with the EARLINET work protocol. During the EARLINET project, DREAM (Dust REgional Atmospheric Model) is used as one of the forecasting models to issue early warning of Saharan dust events over Europe (DREAM:<http://www.bsc.es/projects/earthscience/DREAM>). DREAM- weather forecast maps elaborated by Barcelona Supercomputing Senter gives an image of the wind direction and speed, position of cloud fields and magnitude of dust load in the atmosphere above North Africa and Europe. Also, information about the origin and path that the air-mass passed before their arrival over the lidar site we obtain from the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler R. et al., 2003; Rolph G., 2011). It represents a complete system for computing simple air parcel trajectories to complex dispersion and deposition simulations. The calculations of backward air mass trajectories give a plot of the road that the air mass traversed for a chosen time period before to arrive to the location of lidar observations. Both DREAM and HYSPLIT models are freely available on the Web.

Lidar system and data processing

The Nd:YAG lidar used in the experiments was developed in the Laser Radar Laboratory (Atanaska D. Deleva et al., 2008; A. Deleva et al., 2010). The radiation source is a Q-switched frequency-doubled Nd:YAG laser (pulse energy up to 600 mJ at 1064 nm, 80 mJ at 532 nm; pulse duration 15 ns FWHM; fixed repetition rate 2 Hz; beam divergence 2 mrad). The lidar is mounted on a stable metal coaxial construction allowing reliable fixing and precise synchronized mutual motion (horizontal and vertical) of both the telescope and the output laser beam. The radiation backscattered by atmospheric molecules and aerosols is received by a Cassegrain-type telescope (aperture 35 cm; focal distance 200 cm). The parallel output beam formed by the telescope output optics is passed to a spectrum analyzer for separation of the incoming optical signals. The main lidar elements defining the measurement quality of the system are the photo-receiving modules and the unit

recording the sounding pulses. Each module of Nd:YAG lidar comprises a photo-receiving sensor, an amplifier, a high-voltage power supply, a 14-bit analog-digital converter (ADC), and a Hi-Speed 480 MHz USB 2.0 interface for computer connection. Received signals from the atmosphere are digitized every 100 ns with an ADC, resulting in a 15 m range resolution (about 7.5 m altitude resolution). Thus, the Nd:YAG lidar measures the temporal evolution of the aerosol field with high time and height resolution. The data acquisition software for calculating the atmospheric backscatter coefficient profiles is based on the widely used Klett-Fernald-Sasano algorithm (J. D. Klett, 1981; F. G. Fernald, 1984; Ya Sasano et al., 1985). The magnitude of this coefficient value is proportional to the aerosol density so the retrieved lidar vertical profile illustrates the stratification of the aerosol loading over the lidar station. We present here the results mainly in terms of vertical atmospheric backscatter profiles (x-axis represents the value of the calculated atmospheric backscatter coefficient; y-axis – the altitude above sea level, ASL). The measurement date and laser sounding wavelength are written over the lidar profile plot. Also in the paper we include the range-corrected signals (RCS) maps in order to display the temporal evolution of the aerosol loading of the atmosphere. RCS is produced by subtracting the estimated background noise from the raw lidar signal and multiplying by the square of the distance to the backscattering atmospheric sample.

The Nd:YAG lidar is configured in a mono-static biaxial alignment pointing at a slope angle of 32° with respect to the horizon, as determined by its position in the laboratory. Thus, although signals from as far as 30 km are being recorded, the maximum sounding height is limited to 16.4 km. The good parameters of all the laser, telescope, photo-receiving modules and software make it possible for the Nd:YAG lidar to be utilized for carrying out fast remote measurements of the atmosphere from 130 m above ground level (AGL) (approximately 700 m ASL) to the tropopause.

Lidar observations and discussions

In the end of May an alert was distributed to all EARLINET stations because on 21 May 2011 the Iceland's most active volcano, Grimsvötn, erupted, sending a plume of ash and steam to about 12-14 km high into the atmosphere and causing disruption to air travel in North-Western Europe. The eruptions ceased on 28 May 2011. The volcanic ash propagated quickly in the atmosphere traversing most of European countries. Its trajectories were forecasted by Navy Aerosol Analysis and Prediction System (NAAPS) (NAAP: <http://www.nrlmry.navy.mil/aerosol>) and observed by many meteorological stations, to prevent unintended consequences of the airplane transport. During Grimsvötn eruption all lidar stations of the EARLINET network performed a large campaign of observations to identify the location, height and thickness of volcanic aerosols transported in the air over Europe. As a partner in EARLINET project, Sofia lidar station accomplished monitoring of the atmosphere in the period 26-30 May, regardless that NAAPS didn't forecast that the atmosphere over Bulgaria will be polluted by volcanic aerosols. The experimental results from those investigations we describe by the vertical profiles of the atmospheric backscatter coefficient in Fig.1 and Fig.3. The height of the profiles on Fig.1 was limited to 5-6 km. because the atmosphere remained clear at higher altitudes and we didn't register neither

clouds nor aerosol layers. The decrease of the profile height makes it easier to visualize the mass stratification of the boundary aerosol layer with thickness of 3-4 km. The aerosol profiles show relatively large values of the atmospheric backscatter coefficient in the range of $2-3 \cdot 10^{-6} \text{ m}^{-1} \text{ sr}^{-1}$ for altitude 1.5-2.5 km. It can be assumed that the reason for this is not only the higher density of the aerosol particles at that altitude, but also the higher humidity of the air, as for the days of the investigations Barcelona Supercomputing Center (BSC) forecasted cloud cover over Bulgaria (the upper part of DREAM maps on Fig.2).

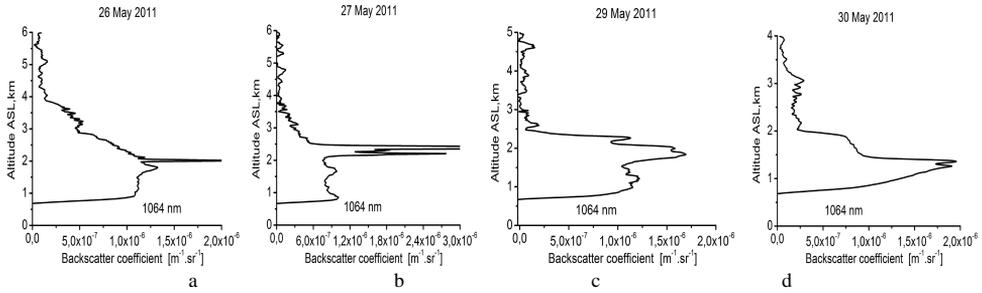


Fig.1. Lidar monitoring of the atmosphere performed on 26, 27, 29 and 30 June 2011. Atmospheric backscatter coefficient profiles retrieved on the basis of the lidar measurement data.

Regarding the nature of the particles in the observed thick aerosol layer immediately above the earth surface we could assume that they are either Saharan dust or have anthropogenic origin. The Balkans are located in the south-east part of Europe, thus continental type aerosols are dominant with industrial or transport influences. However, because they are situated in the eastern Mediterranean basin, the most relevant in the characterization of aerosol properties is the African-desert dust which distorts the atmospheric composition. We have to discard the possibility that in the end of May 2011 in the atmosphere over Sofia there were desert aerosols, because the lower part of the DREAM maps on Fig.2 and Fig.3 shows that Saharan-dust transport over Bulgaria is not expected for the measurement period 26-30 May.

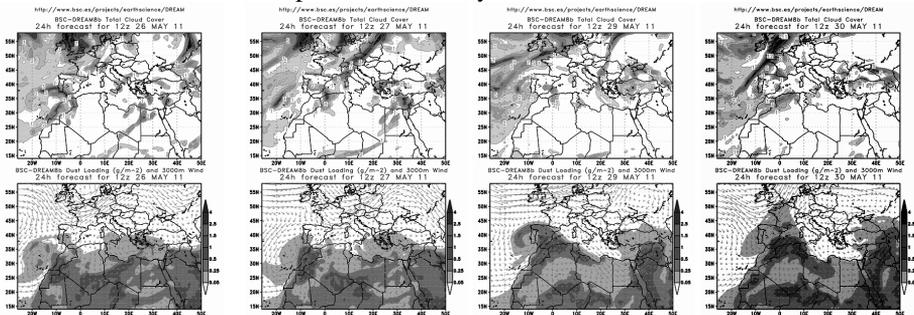


Fig.2. Forecast maps provided by Barcelona Supercomputing Center (BSC) showing no-Saharan-dust transport over Bulgaria for the days of lidar observations (26, 27, 29 and 30 June 2011).

The results of the investigation on 28 May are shown on a separate figure (Fig.3). In that day except for the low-situated stable aerosol layer, observed the whole week above the city, for a short period (about 1 hour) we registered additionally Cirrus cloud in the

altitude range 9-11 km. Cirrus clouds are formed in the upper levels of the troposphere at heights greater than 6 km. As a general rule, Cirrus clouds are thin enough to be transparent or very close to it, because humidity is low at such high altitudes (Prabhakara C., 1993, S. Veerabuthiran, 2004; Satyanarayana, M., 2008). Also, Cirrus clouds sometimes have a short lifetime. The RCS-map (Fig.3.c) illustrates that natural characteristic of theirs. It is observed that in the beginning of the monitoring Cirrus cloud was practically transparent, then it became denser, cloud's base lowered to about 9 km and after that Cirrus cloud got totally dissolved.

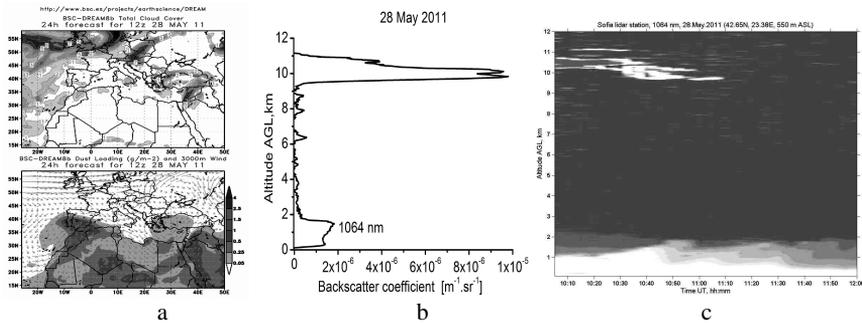


Fig.3. Lidar measurement performed on 28 May 2011. Forecast map provided by Barcelona Supercomputing Center (BCS) showing no-Saharan-dust transport over Bulgaria (a). Backscatter coefficient profile and RCS-map of the time evolution of the aerosol load (b, c).

The most important conclusion from the described investigations is that the atmosphere above Sofia was not polluted with volcanic aerosols ejected during the Grimsvötn eruptions from 21-28 May 2011. However for the whole week of the lidar investigations the air above the city up to altitude of 3-4 km was continuously loaded with aerosols. Those altitudes exceed the maximum top limit of the well developed planetary boundary layer over Sofia which for summer is estimated to be 1800 m (Ts. T. Evgenieva et al., 2009). The planetary boundary layer (PBL) is the lowest layer of the atmosphere, where practically most of the aerosols originating from human activities are situated. Its top limit marks the height of the convection process due to the diurnal cycle of warming and cooling of the Earth surface. The air quality over an urban area depends on the solar radiation reaching the PBL, local pollution sources and processes of vertical mixing and advection. Sofia is situated in a valley surrounded by mountains. Sometimes local meteorological conditions create inversion of the temperature in altitude, which on the other hand suppresses the air circulation. As neither other source of aerosols, nor dust transport from Sahara over the Balkans was forecasted in the end of May 2011, our conclusion was that the smog of human activities and traffic in town caused the observed relatively thick aerosol aggregation.

Barcelona Supercomputing Center forecasted Saharan dust transport above Bulgaria for the period 7-9 June, 2011. We performed lidar measurements every day and we show the results by the atmospheric backscatter profiles on Fig.4. Three profiles are retrieved each averaged for 30 min interval (corresponding to data accumulation of 3600 different raw profiles received at each laser pulse).

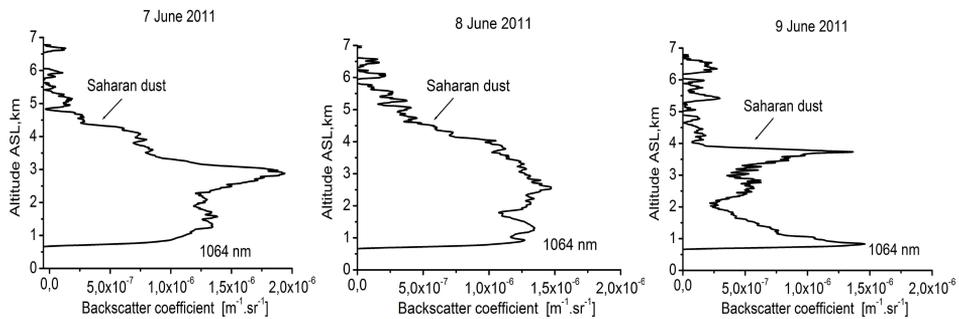


Fig.4. Atmospheric backscatter coefficient profiles, retrieved on the base of the lidar measurement data, obtained during Saharan dust incursion above Sofia, 7-9 June, 2011.

We were performing the investigations in the three days at time interval 9-12 AM. At that time we have registered the stable persistence of aerosol layers above PBL. During the first two days the layers had a top limit 5-6 km, the center of their mass was about 2.5 km, the concentration of particles above it gradually decreased with altitude increase and the border of layers with PBL (about 1.5 km) was expressed mildly. On 9 June we registered change in the aerosol stratification above PBL. The top limit had lowered to 4 km and at that altitude the layer was most dense.

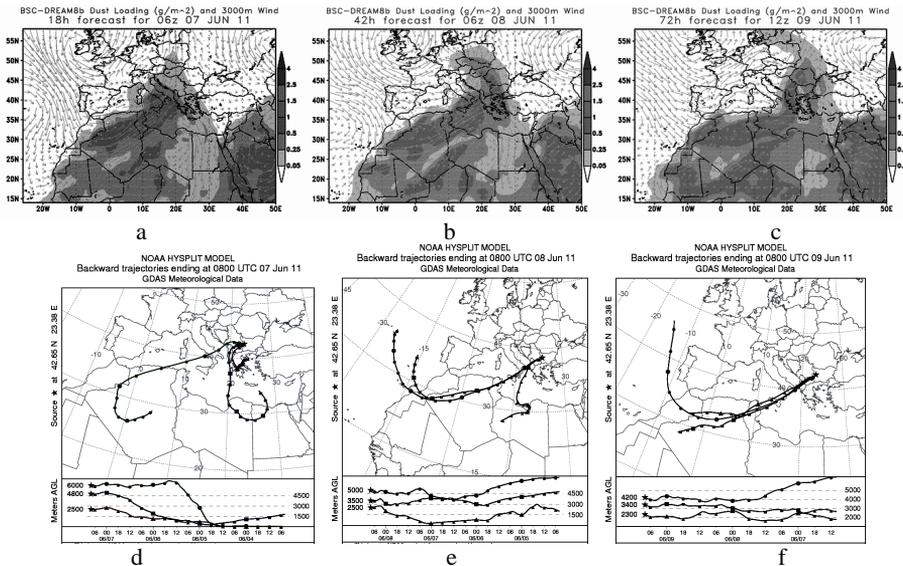


Fig.5. DREAM-forecast maps (a-c), provided by Barcelona Supercomputing Center (BCS) showing Saharan-dust incursion over Bulgaria, 7-9 June, 2011. HYSPLIT-model backward trajectories (d-f) for the days of lidar observations.

With decrease of altitude the concentration of particles decreased sharply and reached its lowest value just above PBL, where the border between the two layers (at altitude 2 km) is highly pronounced. We used information provided by DREAM and HYSPLIT models in order to make conclusions about the origin of the aerosols observed by the lidar. The

corresponding forecast maps are shown on Fig.5. The DREAM maps (Fig.5.a-c) show that dense Saharan dust covered the area of Bulgaria. For the measurement days we calculated HYSPLIT backward trajectories for 100 hrs duration in the altitude range 1.5-6 km. The black-and-white image makes hard distinguishing the trajectories in altitude, so we will clarify it in more detail. It is necessary especially for 7 June (Fig.5 d). The trajectories in the altitude range 1.5-2.5 km (on Fig.5.d is include one of them at 2.5 km only) start over close to Bulgaria regions, which atmosphere was loaded with Saharan dust. It is visible on the lower part of the HYSPLIT map that the air masses which during the measurement were above Sofia at altitude 2.5 km before have moved directly above the Earth surface. Therefore we infer that the air above Sofia up to height of 2.5 km contained substantial amount of anthropogenic aerosol and ones emitted by the Earth surface. The higher calculated trajectories up to 6 km start over African desert regions or near the Sahara surface, illustrated by the included trajectories at 4.8 and 6 km (Fig.5 d). The HYSPLIT information leads us to the conclusion that the air masses in the upper part (3-5 km) of the registered aerosol layer consisted mainly of desert aerosols. For the remaining two days (8 and 9 June) it is not necessary to describe the backward trajectories in the altitude range 2.3-5 km because it is obvious that they pass over Sahara desert and across the highly dusted space over Mediterranean Sea before the end point above Sofia. All information about aerosol layer origin, its altitude above ground, persistence during lidar observations, confirmed the conclusion of a long-distance Sahara dust transport above Sofia from 7 to 9 June 2011 in the altitude range 2-5 km, where we most often register desert aerosols

Up to the end of June Barcelona Supercomputing Center didn't forecast other Saharan dust outbreaks over Bulgaria. But EARLINET community was again mobilized in the last days of the month because in some lidar stations in Italy and Germany were registered a layer at 16 km and a weaker layer at about 12 km.

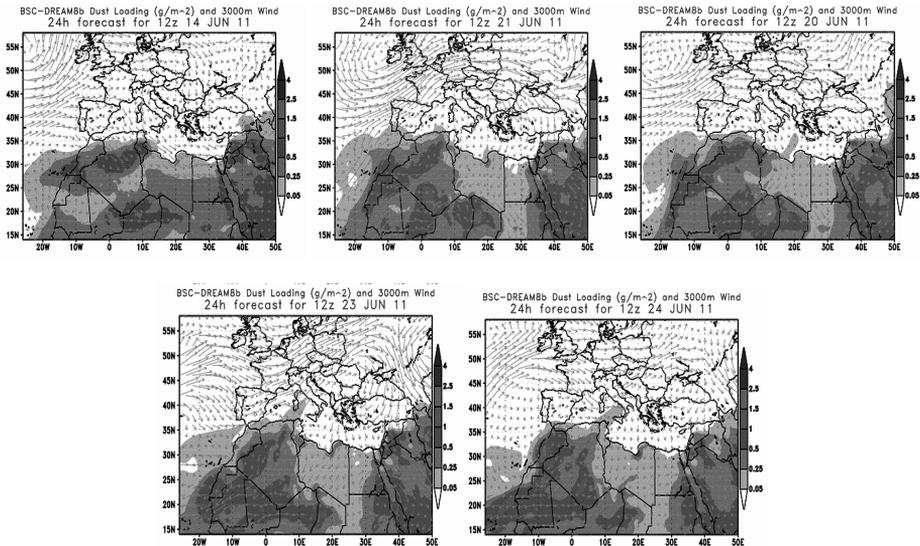


Fig.6. Forecast maps provided by Barcelona Supercomputing Center (BCS) showing no-Saharan-dust transport over Bulgaria for 14, 20, 21, 23 and 24 June 2011.

It was suspected that it is expected development of sulphate particles 3-4 weeks after the injection of sulphur dioxide from Grimsvötn volcano. The Sofia lidar station performed monitoring of the atmosphere for the period 20-24 June due to the same reason. We can say beforehand that those days we didn't register any layers in the upper troposphere, until again in the end of May we observed exceptionally rare aerosol load of the air above Sofia. We could conclude that it has anthropogenic origin because there is no information for other sources of aerosols like fires, volcano eruption or desert dust transport for the end of June. DREAM forecasts (Fig.6) for the days of the lidar monitoring show an atmosphere free of desert dust over Balkans. On Fig.7 are shown lidar results obtained on 14, 20, 23 and 24 June 2011. We have observed during these days aerosol load above Sofia up to 3-6 km heights. All the measurements were performed in the interval around noon (9 AM – 13 PM) and only on 23 June there is additional one around Sunset. For that day we present profiles from the daily as well as from the evening monitoring, because they show the daily cycle in urban aerosols caused by warming the ground (Massimo Del Guasta, 2002). The solar radiation reaching PBL is the reason for its daily changes from the state of stable residual layer (about Sunrise) to well developed mixing layer (during noon and shortly after). When look at Fig.7 it becomes visible that the profiles from the daily measurements from 20-th (Fig.7.b) and 23-th (Fig.7.d) June have quite similar contours.

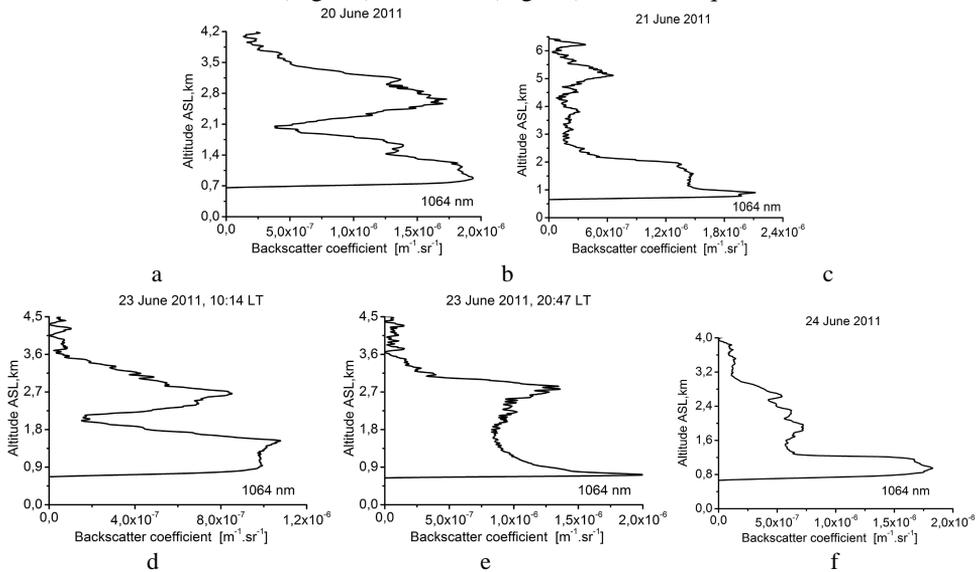


Fig.7. Atmospheric backscatter coefficient profiles retrieved on the basis of the lidar measurement data obtained on 14, 20, 21, 23 and 24 June 2011.

That leads to the conclusion that the aerosol layers registered during the two days have had slightly different mass altitude stratification. Taking into consideration that during Summer both the residual layer as well as the mixing layer over Sofia could raise to 1800 m height (Ts. T. Evgenieva et al., 2009), then our explanation of the received results is that for both days we have observed typical for the Summer residual layer with thickness about 1800 m and above it another one with top limit 4 km and 3.6 km, respectively. As a result of the hot

weather the fine aerosols could possibly have risen high above PBL the day before and have stayed there longer as a result of the weaker gravity and the absence of wind.

The comparison of the profiles from the daily (Fig.7.d) and the evening (Fig.7.e) measurement on 23th June shows that the registered in the morning two clearly defined layers completely merged, but the top limit of the aerosol load remains constant with value of 3.6 km. The merging of the layers is caused mainly by the convective processes in PBL. Every day they start before the Sunset and their intensity during the day depends on the change of the temperature of the air. The convective and adjective processes in PBL lead to the mixing of the particles in the layer close to the Earth surface and to the formation of well defined mixing layer during noon. The remaining profiles on Fig.7 show that on 20, 21 and 24 June we have registered aerosols up to 4-6 km. Besides that in the lower part of the layers we have calculated high values for the atmospheric backscatter coefficient reaching $2 \cdot 10^{-6} \text{ m}^{-1} \text{ sr}^{-1}$. We assumed that it is caused by water vapor flooding of small anthropogenic particles during sunrise.

It is disturbing that, similarly to May and June, the thick urban aerosol layers over Sofia have been also registered during the next warm/hot months. It was imperceptible by eye, but clearly noticeable for the laser light at 1064 nm wavelength. Some atmospheric backscatter profiles retrieved from the measurement data obtained in July and August 2011 we present on Fig.9. The investigations were carried out in the morning. The DREAM maps (Fig.8) show that for the corresponding days there was no Saharan dust transport over Bulgaria.

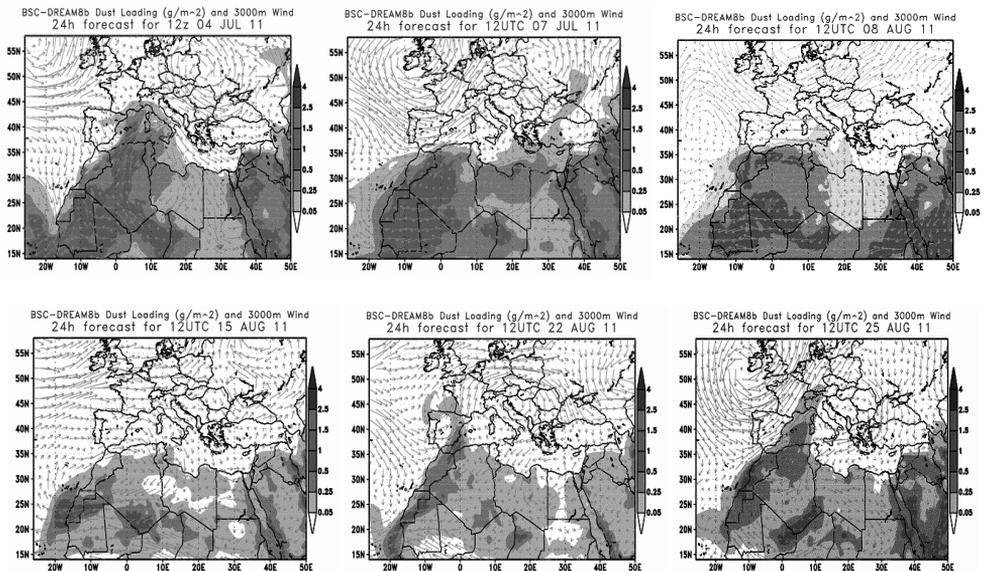


Fig.8. Forecast maps provided by Barcelona Supercomputing Center (BSC) showing no-Saharan-dust transport over Bulgaria for 14, 20-the days of lidar observations.

The lidar profiles on Fig.9 show the unsubstantial difference in the mass stratification of the registered aerosol layers. The main result of the measurements is that the atmosphere over Sofia in heights 3.5-4 km have been again loaded with urban aerosols.

The heat and the lack of winds in the valley, where Sofia is located, caused elevation of the city smog to such unusual heights.

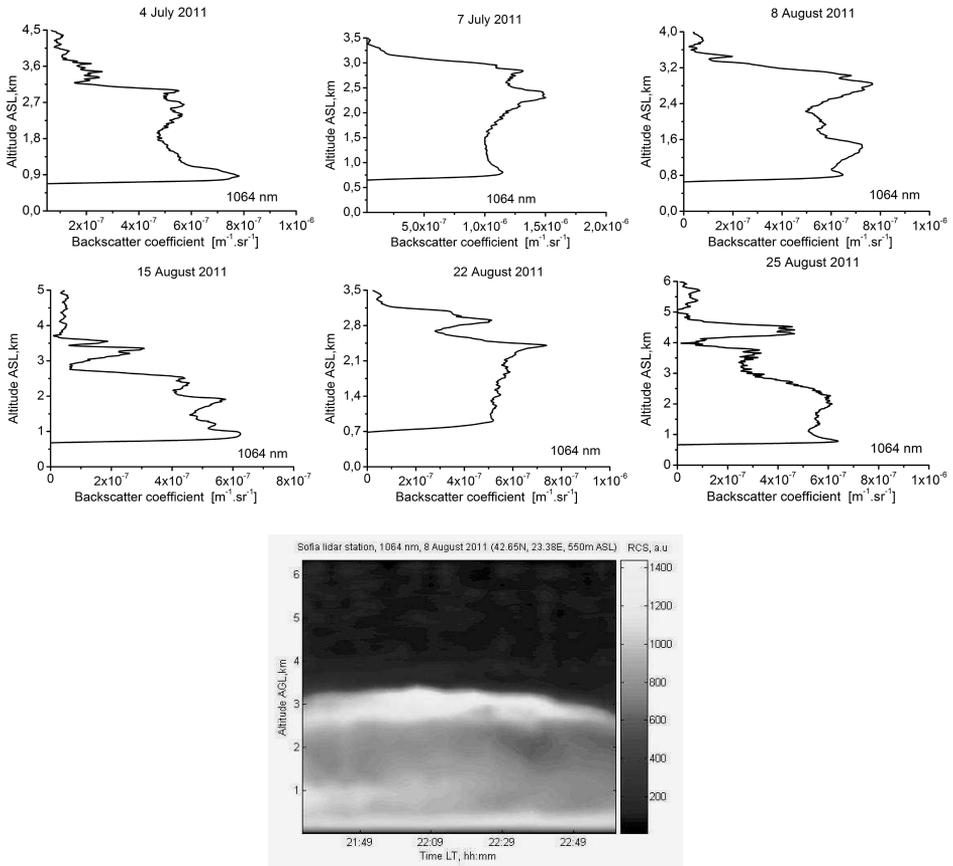


Fig.9. Atmospheric backscatter coefficient profiles retrieved on the basis of the lidar measurement data obtained in July and August 2011. RCS-map of the time evolution of the aerosol load up to 3.5 km observed on 8 August 2011.

The analysis of the data from the separate measurements shows that pollution is permanently present in the air above the city. As an example of this on Fig.9 we present RCS-map of the time evolution of the urban load observed on 8 August 2011. RCS-map illustrates relatively stable atmospheric aerosol stratification and its invariability during the lidar observation.

Conclusions

Lidar investigations of the atmosphere, performed in the period May-August 2011 are described. The experimental results could be summarized as follows. In the end of May the air above Sofia was not polluted by volcanic ash ejected during the Grimsvötn eruptions

(21-28 May 2011). In the period 7-9 June a long-distance Sahara dust transport above Sofia was detected. The most important conclusion concerning the air quality is that during most of the hot-month days in 2011 the atmosphere had been persistently loaded by anthropogenic aerosols at a height of 3.5-4.5 km above the city.

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Лидарни изследвания на тропосферата, направени през лятната 2011 измерителна кампания

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Резюме. В тази работа ние описваме лидарни изследвания на тропосферата, направени от май до август месец на 2011. През този период са извършвани както регулярни седмични EARLINET измервания така и всекидневни лидарни наблюдения на атмосферата повреме на избухванията на Grimsvötn вулкана в Исландия и при пренос на прах от пустинята Сахара. Лидарният мониторинг е направен с аерозолен лидар с Nd:YAG лазер. Експерименталните резултати са представени чрез вертикални профили на коефициента на обратно разсейване на атмосферата и цветни карти на времевата еволюция на аерозолната стратификация. За да направим изводи за произхода на частиците в регистрираните аерозолни слоеве ние изчислявахме HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) обратни траектории и използвахме DREAM (Dust REgional Atmospheric Model) прогнозите за дните на измерванията. Бяха направени изводи, че атмосферата над София не е била замърсена с вулканична пепел повреме на избухванията на Grimsvötn вулкана (21-28 май 2011). Най-важното заключение, което е от значение за качеството на въздуха над София, е, че през преобладаваща част от дните на горещите месеци през 2011г. над града до височина 3.5-4.5 км атмосферата е била трайно натоварена с антропогенни аерозоли. Описаните експерименти са част от регулярните лидарни изследвания на атмосферата, които извършваме в рамките на Европейския проект EARLINET.