Lidar detection and characterization of multi-type aerosol layers in the troposphere above Sofia, Bulgaria

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Abstract. Aerosols can directly and strongly affect the atmospheric energy budget, the climatic processes, the ecological state of the environment, and the biosphere and human health. This is why the detection and identification of aerosols in terms of type and origin and their optical and microphysical characterization are of great scientific and practical importance. Lidars are a widely recognized and proven tool for detecting and studying atmospheric aerosols. In this paper, we present results of lidar detection and characterization of aerosol layers present in the troposphere over the city of Sofia. The measurements were performed at the wavelengths of 1064 nm and 532 nm by using the Nd:YAG-laser based lidar of Sofia lidar station, part of the European Aerosol Research Lidar Network (EARLINET) and the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS). Time-averaged vertical profiles of the aerosol backscatter coefficient and the backscatter-related Ångström exponent were retrieved and analyzed. Color-coded height-time diagrams of the vertical mass distribution in the observed layers and its temporal dynamics were also obtained. Based on information from forecast models, it is concluded that the aerosol layers registered contained marine aerosols, forest fire smoke, indirectly and directly transported dust from Sahara, as well as continental, regional and local aerosols from different sources.

1. Introduction
The atmospheric aerosol (a mixture of tiny solid and liquid particles of natural and anthropogenic origin) has a strong impact on the environment and influences life on Earth in different aspects. Global aerosol effects are very difficult to assess because their location in the atmosphere and concentration are constantly changing over time and space. Many studies have been conducted worldwide in the last decades in order to fully understand and characterize in detail the aerosol properties. For that purpose, LIDARs (Light Detection And Ranging) have been proven as efficient tools for studying the aerosol load of the atmosphere and for retrieving the vertical profiles of the aerosol optical properties, as they allow real-time remote measurements with a high temporal and spatial resolution.

The European Aerosol Research Lidar Network (EARLINET, [1]) and the Aerosol Robotic Network (AERONET, [2]) have measured aerosol properties for more than two decades. Recently, the two networks have joined their efforts within the framework of the Aerosols, Clouds, and Trace gases Research InfraStructure (ACTRIS, [3]) [4-6]. The Sofia lidar station (at the Laser Radars Laboratory of the Institute of Electronics, Bulgarian Academy of Sciences) has been involved in coordinated EARLINET measurements since 2002 and is currently part of ACTRIS.
In this paper, we describe remote investigations of the atmosphere over the city of Sofia, Bulgaria, performed on 30 May 2019 and 9 August 2019 using an elastic backscatter lidar based on a Nd:YAG-laser. The lidar’s construction and operation, as well as the lidar data processing, visualization and analysis, have been described in detail in previous publications [7, 8].

2. Results and discussion
Below we analyze two cases of lidar investigation of the atmosphere, in which aerosols are registered from the Earth’s surface up to heights of 4-5 km above ground level (AGL). Hereafter, the quoted values of elevation (height or altitude) are as measured AGL.

The lidar measurements under consideration were carried out on days for which the Barcelona Supercomputing Center [9, 10] forecasts did not anticipate Saharan dust intrusions over most of Europe, including Bulgaria (figure 1; Bulgaria’s location is indicated by a red circle).

| Figure 1. NMMB/BSC-Dust model forecast maps for 30 May 2019 (a) and 9 August 2019 (b). |
|---------------------------------------------------------------|
| (a)                                                          |
| (b)                                                          |

In figure 2, the pre-processed lidar data recorded at the wavelength of 1064 nm are presented as range-corrected signal (RCS) color maps in height-time coordinates consisting of individual profiles with time resolution of 1 min (a) and 3 min (b), to illustrate the temporal dynamics of the air mass distribution during the two lidar measurements. The RCS profiles are smoothed over the height range 2-5 km using a moving average filter with a window evenly increasing from 45 m to 150 m, in order to suppress the noise oscillations, thus distinguishing more clearly and visualizing better the aerosol-containing atmospheric domains.

| Figure 2. RCS-color map of the temporal evolution of the aerosol-density above Sofia, as measured at the wavelength of 1064 nm on 30 May 2019 (a) and 9 August 2019 (b). |
|---------------------------------------------------------------|
| (a)                                                          |
| (b)                                                          |
The color maps reveal the presence of aerosols up to heights of approximately 3.5-4 km (figure 2 (a)) and 4-4.5 km (figure 2 (b)). These altitudes exceed the upper limit of the well-developed atmospheric boundary layer (ABL) over Sofia (estimated to be 1.8-2 km in summer). Therefore, the detected aerosol loads above 2 km are very likely of non-local origin, because practically most of the aerosols originating from local emission sources and human activities are normally confined within the ABL.

From the coloring of the map in figure 2 (a), one can conclude that dynamic processes had taken place in the lower troposphere above Sofia on 30 May 2019, which had led to mixing and inhomogeneous distribution of the aerosol density up to a height of 3.5 km. In contrast, on 9 August 2019 (figure 2 (b)), layers with nearly homogeneous structure and well-defined boundaries were registered, indicating a calmer atmosphere with virtually no convective processes. The lidar-observed aerosol mass stratification will be described in more detail below, where we analyze separately the two cases considered. For each of them, we apply a set of figure panels designated as (a), (b) and (c), displaying:

(a) Time-averaged lidar profiles of the atmospheric aerosol backscattering coefficient (BSC) at 532 nm and 1064 nm retrieved by using the widely adopted inversion approach of Klett-Fernald [11, 12] in its version assuming a constant lidar ratio (the ratio of the extinction coefficient to the backscattering coefficient).

(b) HYSDPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model backward trajectories by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory’s (ARL) [13, 14], which yield plots of the path traveled by the air masses in a chosen time period (in hours) before reaching Sofia during the measurements;

(c) Vertical profiles of the backscatter-related Ångström exponent [15] obtained from the two BSC profiles (registered at 532 nm and 1064 nm), whose value is regarded as an indicator of the dominant particle size fraction in the aerosol layers.

In retrieving the aerosol BSC profiles, a constant lidar ratio of 50 is assumed, a value commonly chosen when heterogeneous aerosols are detected, as in the case considered here. The altitude range of the profile calibration in which reference values of BSC are defined is 6-8 km (being as a rule at heights in the free troposphere where the aerosol content is negligible compared to the molecular one).

Figure 3. Lidar monitoring performed on 30 May 2019: (a) Retrieved vertical atmospheric backscattering coefficient profiles obtained at 532 nm and 1064 nm; (b) HYSDPLIT-backward trajectories ending above Sofia during the measurements; (c) Backscatter-related Ångström exponent vertical profile.

The lidar profiles in figure 3 (a) reveal that on 30 May 2019 the main aerosol mass was located in the altitude range 0.5-3.3 km. The particles were distributed in the layer relatively evenly. Upwards, their concentration gradually decreased to a height of about 3.8 km. Then, a well-defined layer with
boundaries at 3.8-4.8 km was registered, in which the density of the particles did not change significantly in height and was much smaller than that in the lower main layer.

Figure 3 (b) shows three of the HYSPLIT backward trajectories calculated for a time duration of 310 hours before the lidar measurement on 30 May. They end above Sofia at altitudes of 1000 m, 2200 m and 3700 m, which fall near the center and borders of the main aerosol layer registered. The path of the air masses that they outline is similar to that displayed by other trajectories calculated for different altitudes in the range 0.5-5 km. The trajectory analysis shows that the air masses had moved over the Atlantic Ocean for a quite long time before ending over the city of Sofia. Some of them had passed very close to the water surface, which is illustrated by the red trajectory on figure 3 (b). This suggests that they likely carried marine aerosols (sea salt) and moisture. Another part of the air masses had been moving over areas of the USA and Canada (green and blue trajectories), where in accordance with the NOAA ARL Smoke Forecasting System (SFS) [16], devastating forest fires raged in May 2019 and, therefore, it is very likely that part of those masses comprised fire smoke. Figure 3 (b) also shows that shortly prior to reaching Sofia, the air masses had crossed the atmosphere over the Mediterranean Sea near North Africa. The forecasts of the NMMB/BSC model for four days before revealed that Saharan dust was permanently present in the atmosphere above the mentioned above areas. This allows us to assume that, among other type of aerosols, the air masses over Sofia during the measurement also contained indirectly transported Saharan dust, i.e. carried by air masses that have not passed above the desert.

Figure 3 (c) shows the calculated vertical profile of BAE, with a mean BAE value of 1.26±0.46. The plateau zone in the altitude range 0.4-3.3 km, corresponding to the main aerosol layer in figure 3 (a), has a mean BAE value of 1.01±0.11, qualitatively indicating moderately coarse aerosol modes. The well-defined two-peak bulge of BAE in the interval 3.3 to 4.8 km, corresponding to the upper aerosol sub-layer (with mean BAE of 1.72±0.56) and the layer interface in the interval 3.3-3.9 km (with mean BAE of 1.61±0.38), indicate rather finer aerosol fractions. Based on the analysis of HYSPLIT backward trajectories, we attribute the lower BAE values in the main aerosol layer to continental and marine aerosols. In addition, it is likely that the upper sublayer contained fire smoke from USA and Canada, in correlation with the high BAE values reached (up to 2.5), typical for fine smoke aerosols, especially after long-range transport, as in the current case.

The set of figures relating to the measurement performed on 9 August 2019 is presented below.

Figure 4. Lidar monitoring performed on 9 August 2019: (a) Retrieved vertical atmospheric backscattering coefficient profiles obtained at 532 nm and 1064 nm; (b) HYSPLIT-backward trajectories ending above Sofia during the measurements; (c) Backscatter-related Ångström exponent vertical profile.

The time-averaged lidar profiles of figure 4 (a), as well as the color map of figure 2 (a), show that on 9 August 2019 three distinct layers were registered. The lowest of them up to a height of about 1.7-
1.8 km (nearly coinciding with the seasonal ABL above Sofia) is the densest one and has a pronounced center of mass at 1.2 km. The following higher layers are less dense and have upper limits of approximately 3.2 km and 4.7 km.

Selected HYSPLIT trajectories calculated for 370 hour duration and falling into the registered three layers are included in figure 4 (b). Based on the trajectory analysis, one could conclude that during the lidar measurements in the atmosphere up to altitude of 5 km, both general and specific aerosols were present in the layers registered. In general, air masses up to 5 km had passed over areas of the Atlantic Ocean and Europe, some of which had been loaded with Saharan dust. Therefore, one can assume that they comprised certain amounts of marine aerosols, indirectly transported desert dust and continental aerosols of natural and/or anthropogenic origin. The distinctive feature of the low trajectory is that its initial part passes close to the water surface of the Atlantic Ocean and it is very likely that in the ABL particles of sea salts were present that were larger than the usual local aerosols. As shown in figure 4 (b), air masses at an altitude of 2.8 km passed over the Eastern parts of the United States, where forest fires raged in many areas in the late July and August. It is worth noting that for the measurement period, approximately 17:00−19:00 UTC, we calculated a number of trajectories ending above Sofia in the altitude range 1.8−4.6 km, which had passed over large areas of the US territory. Therefore, it is very likely that there was smoke in the registered aerosol layer up to 5 km, among other aerosol types. The uppermost trajectory reveals that the air masses were moving at an altitude of 7 km above Saharan regions covered by dust. Given that the ABL of Sahara reaches a height of 5−6 km, it can be argued that desert dust could be present in the upper registered layer above Sofia.

Figure 4 (c) shows the retrieved vertical profile of BAE with a mean BAE value of 1.25±0.45. In the densest bottom aerosol layer, the BAE varies from 0.67 to 1.24, with a mean value of 0.96±0.19, showing domination of moderately coarse aerosol fractions. The intermediate layer from 1.8 km to 2.75 km has an average BAE value of 0.98±0.1. The uppermost aerosol layers in the range 2.8−5 km have an average BAE value of 1.54±0.48. Up to 4 km, the BAE varies slightly in the range 1−1.3, while in the range 4−5 km it increases from 1.21 to 2.52 with a mean value of 1.96±0.42, thus indicating a clear domination of fine aerosol fractions. Combining the NOAA HYSPLIT and the ARL SFS data, the aerosols in the lower profile parts were determined as possibly being of marine, urban and general origin, whereas those above 4 km were identified as likely containing fire smoke from wildfires raging in the USA in the end of July 2019.

3. Conclusions
The atmospheric lidar studies described in this work were performed on days without direct Saharan dust intrusions over Bulgaria. Aerosol loading of the atmosphere up to heights of 4.5 km was observed, affecting the air quality above the city of Sofia. Inferences about the origin and type of aerosols were made using air transport modeling and forecasting data from the NMMB/BSC-Dust and HYSPLIT models. It was concluded that the relatively thick aerosol layers registered contained multi-type particles of different size, such as marine aerosols, forest fire smoke, indirectly and directly transported dust from Sahara, as well as continental, regional and local aerosols from different sources.

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