Spring-to-summer observations of the aerosol load over Sofia-city using near-horizontal lidar sensing

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Abstract. Using lidar and contact facilities, the relation is investigated experimentally between the extinction coefficient and the mass concentration of near-ground aerosol pollution over some districts in Sofia City. The extinction coefficient profiles of the aerosol ensembles along the lidar line of sight (LOS) are measured at the lidar wavelengths of 510.6 nm (CuBr-vapor laser source) and 1064 nm and 532 nm (Nd:YAG laser source). The particulate mass concentration is determined using data obtained concurrently by air-quality measuring sensors close to the LOS. Thus, the results obtained of the aerosol mass concentration-to-extinction calibration constant of the lidar concern mainly the sites around the nearest stations. To help the interpretation of the results, the accompanying weather conditions are also described. The experiments are conducted in the spring and the summer and the results obtained reflect the ecological peculiarities of the corresponding city regions of interest. The values found of the calibration constant are consistent with those obtained earlier in the same seasons.

1. Introduction
Atmospheric aerosols of natural or anthropogenic origin have a significant impact on climatic phenomena and processes on a global and regional scale [1]. In the near-surface atmospheric layers, they have a strong and direct effect on the air quality, local ecosystems and human health. For these reasons, studying and monitoring the aerosol distribution, concentration and dynamics close to the ground, as well as localizing the aerosol contamination sources and identifying the aerosols by type and origin, are of great scientific and societal importance, especially over densely populated regions [2-5].

The distribution and temporal evolution of the near-surface aerosol field has long been a subject of particular attention of the Laser Radar Laboratory at the Institute of Electronics of the Bulgarian Academy of Sciences (IE-BAS) [2, 5]. In this work, using data from near-horizontal lidar remote sensing and contact probing, an investigation is performed of important optical and physical characteristics of the aerosol pollutants over ecologically specific regions in Sofia. These characteristics are the extinction coefficient of the near-ground aerosol ensembles, their mass concentration and the mass concentration-to-extinction ratio. The last value mentioned is actually the calibration constant of the lidar with respect to the aerosol mass concentration in the populated urban zone. The knowledge of this constant allows

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one to determine by lidar the variable spatial distribution of the particulate matter (PM) mass concentration and to predict potential negative effects of the PM pollution on the human health.

The investigation performed follows some main goals, including: determination by lidar of the near-ground distribution of the aerosol extinction coefficient along some chosen (near) horizontal line of sight (LOS); ongoing tracking and processing of data from the existing in-situ-probing ecological stations providing versatile information of the aerosol pollutants, including their mass concentration; estimating the lidar mass concentration-to-extinction calibration coefficient at concrete radiation wavelengths; current tracking of the weather conditions; interpretation of the results obtained, comparing them with each other and with earlier results obtained in spring and summer for Sofia region [6].

2. Experimental approaches, instruments, and conditions

2.1. Lidar systems and observation zones

The near-horizontal lidar sensing was conducted using two lidar systems developed in the Laser Radar Laboratory of the IE-BAS. These systems are of monostatic non-coaxial type. They are based on the use as sources of sensing radiation of a pulsed CuBr-vapor laser emitting the wavelength of $\lambda = 510.6$ nm (figure 1 (a), inset), and a frequency-doubled Nd:YAG laser emitting the fundamental wavelength $\lambda = 1064$ nm and the first harmonic 532 nm (figure 1 (b), inset). A detailed description of both lidar systems is given in Refs. 2 and 5. They are installed, respectively, on the roof and on the fourth floor of the IE-BAS building (42.65N, 23.38E; 590 m a.s.l.) in the southeast part of Sofia. During the experiments, the LOS of the CuBr-laser-based lidar system was directed northwest, toward the Fakulteta district (figure 1 (a)) located at a distance of 12 km from IE-BAS. The LOS of the Nd:YAG-laser-based lidar system is directed southwest, toward Vitosha Mountain (figure 1(b)). The in-situ measured PM$_{10}$ mass concentrations that are necessary for calibrating the first lidar are provided by the air-quality measuring sensors installed in different Sofia districts in 2019 in the framework of the AIRTINGS project [7]. The data concerning the second lidar are provided by PM measuring sensors part of the Citizen Science project Luftdaten.info [8]. The position of the LOS sensors closest to the lidars are indicated in figures 1 (a) and 1 (b).

![Figure 1. Photographs of the CuBr-laser-based lidar (a) and the Nd:YAG-laser-based lidar (b) LOS and the position of the closest air-quality sensors on the map of Sofia City.](image)

2.2. Determining the aerosol extinction and backscatter coefficients and the mass concentration

The lidar signal profile $P(r)$ as a function of the lidar parameters and the LOS profiles of the atmospheric extinction $\sigma(r) = \sigma_a(r) + \sigma_m(r)$ and backscattering $\beta(r) = \beta_a(r) + \beta_m(r)$ coefficients is described by the so-called lidar equation [9]; $\sigma_a(r)$ and $\sigma_m(r)$ and $\beta_a(r)$ and $\beta_m(r)$ are, respectively, the aerosol and molecular extinction and backscatter coefficients. The solution of the lidar equation with respect to $\sigma_a(r)$ and $\beta_a(r)$, provided that $\sigma_m(r)$ and $\beta_m(r)$ are known, is obtainable using the Klett-Fernald or the slope (logarithmic-derivative) approaches [9-11]; $r = ct/2$ is the coordinate along the LOS, and $t$ is the time after the emission
of the sensing laser pulse. The first approach leads to a stable solution of the lidar equation and is being widely applied now. In the case of horizontal or quasi-horizontal lidar sensing of the near-surface atmospheric layer, there may exist LOS zones with relatively homogeneous aerosol composition and concentration allowing one to employ the slope method for determination of the aerosol extinction coefficient $\sigma_a(r)$. In this case, the extinction coefficient $\sigma(r)$ is expressible through the derivative (the slope) of the natural logarithm of the so-called $S$-function [range-corrected lidar profile (RCLP) or range-corrected signal (RCS)] $S(r) = r^2 P(r)$ as [9]

$$\sigma(r) = -0.5 \frac{d \ln S(r)}{d r}.$$ (1)

In the present experiments, the near-surface aerosol conditions were quite favorable for using the slope method to determine the aerosol extinction at practically all the LOS distances of interest. The LOS distribution of the aerosol mass concentration $C_m$ can be estimated remotely by lidar assuming that it is proportional to the aerosol extinction and backscattering coefficients determined by the lidar. The coefficients of proportionality (calibration constants), $K_\sigma = C_m(r)/\sigma_a(r)$ and $K_\beta = C_m(r)/\beta_a(r)$, are determinable using lidar data for $\sigma_a(r)$ and $\beta_a(r)$ and data for $C_m$ obtained by the above-commented in-situ-probing ecological stations.

3. Results and discussions
We will first present and discuss below results of near-horizontal lidar sensing using the CuBr-laser-based lidar system. The first experiment, performed on 19 May 2020, began at 20:06 Local Time (LT) and lasted 60 minutes. By accumulating the signal every five minutes, we obtained a series of short-term RCLPs allowing us to track the dynamics of the aerosol field along the LOS with a temporal resolution of five minutes. Using RCLPs, a distance-time color map was constructed (figure 2 (a)), displaying the temporal evolution of the aerosol mass distribution during the measurements. The long-

Figure 2. Time evolution color maps of RCLPs measured at 510.6 nm on 19 May (a), 21 July (b) and 22 July (c) 2020.

Figure 3. Natural logarithm of 60-minute (a) and 30-minute (b, c) accumulated RCLPs obtained in the experiments performed on 19 May (a), 21 July (b) and 22 July (c) 2020.
term RCLP obtained by accumulating the signal during the 60-minute period (figure 3 (a)) allow us to estimate the one-hour-averaged profile of the aerosol characteristics along the LOS. Such is the period of data averaging for the aerosol mass concentration in the ecological in-situ stations.

The following experiments, on 21 and 22 July 2020, began at 21:47 and 21:19, respectively, and lasted 30 minutes. In these cases, the short-term profiles were obtained by one-minute signal accumulation (figures 2 (b) and 2 (c)), and the long-term profiles (figures 3 (b) and 3 (c) – by 30-minute signal accumulation.

The PM mass concentration is estimated as an arithmetic mean on the basis of data for PM$_{10}$ concentrations provided by the air-quality measuring sensors shown in figure 1. Such one-hour-averaged data concerning the periods of measurement are given in table 1. The meteorological parameters for every time period considered are given in table 2. They were taken from the National Institute of Meteorology and Hydrology (Bulgaria) [12] and were measured by a station located at 300 m westward from the lidars position.

| Date, Time          | PM$_{10}$ [µg m$^{-3}$] | Sensor Number |
|---------------------|-------------------------|---------------|
|                     | # 1 | # 2 | # 3 | # 4 | # 5 | # 6 | # 7 | # 8 | # 9 |
| 19.5.2020, 20:00-21:00 | 15.01 | 12.31 | 13.26 | 14.16 | 12.17 | 16.04 | 13.55 | 11.52 | 14.00 |
| 21.7.2020, 22:00-23:00 | 15.47 | 18.46 | 22.58 | 18.27 | 13.88 | 16.76 | 16.76 | 16.56 | 25.55 |
| 22.7.2020, 21:00-22:00 | 21.53 | 19.34 | 19.65 | 22.58 | 14.74 | 18.60 | 20.88 | 18.67 | 36.35 |
| 07.8.2020, 12:00-14:00 | 7.64 | 6.91 | 8.78 | 10.3 | # 28864 | # 12474 | # 6197 | # 10821 |

Table 1. Mean hourly values of the PM$_{10}$ concentration measured by the air-quality sensors close to the lidars LOS at the time of lidar measurements.

| Date    | Time | Temperature (°C) | Pressure (hPa) | Wind Direction | Wind Speed (m s$^{-1}$) |
|---------|------|------------------|----------------|----------------|-------------------------|
| 19 May 2020 | 21:00 | 21.2             | 946.7          | SE (140°)       | 2                       |
| 21 July 2020 | 21:00 | 23.3             | 949.9          |                | 0                       |
| 22 July 2020 | 21:00 | 25.3             | 948            |                | 0                       |
| 07 Aug 2020   | 12:00 | 24.2             | 946.4          | N (0°)         | 2                       |

Table 2. Weather conditions on the days of measurement.

We first analyzed in detail the results obtained on 19 May. The long-term RCLP is shown in figure 3 (a), where it is seen that the ln of the profile outlines a nearly 12-km-long section of uniform near-ground atmospheric (aerosol plus molecular) extinction. The linear least-squares fit (LLSF) of the profile between 0.6 and 12 km provides a 60-minute-averaged value of the extinction coefficient $\sigma$ of 0.0980 ± 0.0006 km$^{-1}$ (figure 4 (a)). To obtain the aerosol component $\sigma_a$ of the extinction coefficient, one should subtract from the results obtained the value of the molecular extinction $\sigma_m$ for an altitude of 590 m. According to some estimates based on models of mid-latitude summertime atmosphere [9, 13], the value of $\sigma_m$ = 0.0143 km$^{-1}$; i.e., $\sigma_a$ = 0.0837 ± 0.0006 km$^{-1}$. The estimate obtained of the mean mass concentration on the basis of the data in table 1 is $C_m = 13.558 ± 1.438$ µg m$^{-3}$. Using these values of $C_m$ and $\sigma_a$, we obtained the estimate of $K_{\sigma} = 0.162 ± 0.017$ km mg m$^{-3}$. 


The dynamics of the aerosol density was tracked by considering the five-minute averaged profiles. The results show that during the experiment at distances up to 6 – 7 km along the LOS, the near-surface aerosol content is stable and consisting of the same PM having the same extinction coefficient. The aerosol mass concentration should also be uniform at these ranges. At further distances, over 8 – 9 km, the extinction coefficient increases and varies, which is due, perhaps, to the use of solid fuel for heating in Fakulteta district.

![Figure 4. Determination of the slope and the near-ground atmospheric extinction coefficient using LLSF of the logarithmic long-term RCLP presented in figure 3.](image)

The natural logarithm of long-term RCLP obtained on 21 July (figure 3 (b)) outlines a 10-km-long section of uniform near-ground atmospheric extinction, with exception of the initial (near the lidar) transient area and some small abnormal segments along the LOS. The LLSF of the profile between 1.5 km and 3.5 km provides a 30-minute averaged value of the extinction coefficient $\sigma = 0.0876 \pm 0.0006 \text{ km}^{-1}$ (figure 4 (b)). The latter value of the extinction coefficient may be considered as an average over a 7-km-long spatial interval. The estimate of the aerosol component of the extinction coefficient in this case is $\sigma_a = 0.0733 \pm 0.0006 \text{ km}^{-1}$. Note that the quantities concerning the extinction coefficient in July (in the summer) are quite near those obtained in May (in the spring) because of the similar weather conditions. On the basis of the data in table 1, the estimate obtained of $C_m = 18.254 \pm 3.644 \mu \text{g m}^{-3}$. As a result, the estimate of $K_a = 0.249 \pm 0.050 \text{ km mg}^{-1}$.

The long-term RCLP obtained on 22 July is presented in figure 3 (c). It is seen there that, with the exception of the initial (near the lidar) transient area, the $\ln S(r)$ outlines an 11-km-long LOS section of uniform, on the average, atmospheric extinction. The LLSF of the profile between 1.5 km and 6.5 km provides a 30-minute averaged estimate of $\sigma = 0.09245 \pm 0.00056 \text{ km}^{-1}$. The fit with a straight line over the LOS section from 1.5 km to 12 km provides an estimate of $\sigma = 0.0940 \pm 0.0009 \text{ km}^{-1}$ (figure 4 (c)) that is in fact an average over a 30-minute-long time interval and 10.5-km-long spatial interval. The values obtained of the extinction coefficient are close to those obtained on 19 May and 21 July. The estimate of the aerosol component of the extinction coefficient is now $\sigma_a = 0.0797 \pm 0.0009 \text{ km}^{-1}$. On the basis of the data in table 1, we obtain $C_m = 21.371 \pm 6.043 \mu \text{g m}^{-3}$. Finally, using the estimates of $\sigma_a$ and $C_m$ we obtain that $K_a = 0.268 \pm 0.076 \text{ km mg}^{-1}$. This result is statistically consistent within the error limits with the result concerning 21 July. It is consistent as well with the results obtained formerly [6], again in spring and summer, for the coefficient of proportionality between $C_m$ and $\sigma_a$.

Figure 5 presents results from the measurement carried out using the Nd:YAG-laser-based lidar system on 7 August 2020 from 12:00 to 14:00 hours with LOS toward Vitosha Mountain. Distance-time color maps allowing us to track the dynamics of the aerosol field along the LOS with a temporal resolution of three minutes are shown in figures 5 (a) and 5 (d) for the laser wavelengths of 532 nm and 1064 nm, respectively. The corresponding two 120-minute-accumulated lidar profiles are presented in figures 5 (b) and 5 (e).

The LLSF of the natural logarithm of the RCLP for $\lambda = 532$ nm (figure 5 (c)) over the range from 1.25 km to 7.00 km along the LOS provides an estimate of $\sigma = 0.0575 \pm 0.0006 \text{ km}^{-1}$. The corresponding estimate of $\sigma_m$ (see, e.g., [9]) is $\sigma_m = 0.01215 \text{ km}^{-1}$, and $\sigma_a = 0.04535 \pm 0.00060 \text{ km}^{-1}$. The LLSF of the
natural logarithm of the RCLP for $\lambda = 1064$ nm (figure 5 (f)) over the range from 0.5 km to 3.5 km along the LOS provides an estimate of $\sigma = 0.03600 \pm 0.00035$ km$^{-1}$. In this case, $\sigma_m = 0.000759$ km$^{-1}$ [9]. Thus, the estimate of the aerosol extinction coefficient here is $\sigma_a = 0.03524 \pm 0.00035$ km$^{-1}$. The estimate of the mean along the LOS two-hour averaged PM$_{10}$ mass concentration $C_m$ is evaluated using data from the air sensors from the Citizen Science network #28864, 12474, 10821, and 6197 (figure 1 (b)), Table 1. The result obtained is $C_m = 8.4075 \pm 1.545 \mu$g m$^{-3}$.

Figure 5. Time evolution color maps of RCLPs measured on 7 August 2020 at 532 nm (a) and 1064 nm (d); Natural logarithm of the corresponding 120-minutes accumulated RCLPs (b, e); Determination of the slope and the near-ground atmospheric extinction coefficient using LLSF of the logarithmic long-term RCLP at 532 nm (c) and 1064 nm (f).

On the basis of the above-evaluated estimates of $\sigma_a$ and $C_m$, we obtain that $K_e = 0.2386 \pm 0.0439$ km mg m$^{-3}$ at $\lambda = 1064$ nm, and $K_e = 0.185 \pm 0.042$ km mg m$^{-3}$ at $\lambda = 532$ nm. The latter value of $K_e$ is consistent with that obtained on 19 May for $\lambda = 510.6$ nm. In general, a significant volume of monthly and seasonal statistical data is necessary to allow one to establish reliable values of the corresponding specific calibration constants.

4. Conclusions
An interesting and useful observation during the experiments performed in this work was that the specificity of the Sofia city near-surface atmospheric layer is favorable for employing the slope method to determine the aerosol extinction coefficient. That is, there are many large urban areas in the city with homogeneous aerosol content having uniform optical and microphysical properties and aerosol extinction. The results obtained here using the slope method show that the estimates of the extinction coefficients measured in May (in the spring) and July (in the summer) are quite close to each other because of the similar weather conditions and human activities. Expectedly, the estimates of the extinction coefficients measured on 21 and 22 July are practically equal within the error limits. Practically identical are also the estimates of the lidar calibration constants $K_e$ derived from near-surface lidar sensing and contact probing data obtained concurrently on 21 and 22 July 2020. It is interesting to note as well that the estimates of $K_e$ obtained here are consistent, within the error limits, with the values
obtained previously in the spring and the summer of the coefficient of proportionality between the aerosol mass concentration and extinction; the contact probing in this case was performed using aspirator with filters. As a whole, the results obtained in this work show that a careful calibration of the lidars would allow one to use them for express remote monitoring on a vast scale of the distribution of the PM mass concentration. The near-surface urban aerosol conditions are favorable for such a straightforward calibration. Only, a large enough volume of statistical data should be ensured, covering also the autumn and winter months thus allowing one to establish reliable seasonal or annual calibration constants.

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