

VERTICAL VARIABILITY OF AEROSOL ABSORBING PROPERTIES IN THE CARPATHIAN MOUNTAINS.

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Abstract

We present analysis of the vertical variability of aerosol single scattering properties obtained from micro-aethalometer AE-51 mounted onboard of the An unmanned aerial vehicle (UAV) as well as observed by the LB-10 lidar, CHM15k, and CIMEL sun-photometer. Measurements were made in the frame of Poland-AOD network (www.polandaod.pl) at the Radiative Transfer Station SolarAOT in Strzyzow (South-East part of Poland, 49.878° N, 21.861°E, 443 m a.s.l.) between the Sep 2013 and Jun 2014. Vertical profiles obtained from several flights show significant variability of aerosol optical properties with weather conditions such as temperature lapse rate, relative humidity, and wind speed. During intensive smog events in the winter season we found large variability (order of magnitude 10) of the absorption coefficient up to 100-200 m above the valley of small town Strzyzow. This variability can be explain by the mountain-valley circulation during night which leads to accumulation of pollution in the valley. We present sensitivity study of radiative forcing (RF) and radiative heating calculation due vertical variability of absorption coefficient.

1. INTRODUCTION

The climate impact of black carbon (BC) particles originating from fossil fuel combustion or biomass burning, mineral dust and volcanic ash, is still very poorly understood especially its role in the climate system (IPCC, 2013). It is mainly due to two reasons: there are too few observations of BC

vertical profiles and too simplified description of physical processes involving BC used for climate and aerosol modeling (Koch and Del Genio, 2010; IPCC, 2013). BC is reported as the second of the most important anthropogenic climate forcing agent (Ramanathan and Carmichael, 2008; Bond et al., 2013). The averaged top of the atmosphere (TOA) direct RF was estimated by many authors ranging between 0.08 and 1.4 W/m² (Samset et al., 2013; Ramanathan and Carmichael, 2008; Bond, 2013). The wide range of reported RF values is due to different reasons. The most important is related to uncertainty of vertical profile of BC concentration (Samset et al., 2013). Based on extensive results overview Samset et al., (2013), Zarzycki and Bond, (2010) there is a strong need to develop a capability for monitoring of the vertical profiles of the optical properties of absorbing aerosols on a global scale, which will improve our knowledge on their climate impact.

The main motivation for this study is a deficiency of BC profiles existing over Europe (Ferrero et al., 2014) especially in Poland where the aerosol optical depth is relative high. The BC concentration is extremely high during winter (heating season) due to combustion of coal and wood. In the Carpathian Mountains the emission and local circulation lead to increase of BC concentration as well as to horizontal and vertical heterogeneity.

2. METHODOLOGY

This study was done at the Radiative Transfer Station SolarAOT in Strzyzow (South-East part of Poland), 49.878° N, 21.861°E, 443 m a.s.l.) in the Carpathian Mountains (Fig. 1). The SolarAOT station is localized about 200 m above the Valley of Strzyzow in the rural area. The vertical profiles were done during walking trip shows in Fig.1 and

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during flight experiments in Sep 2013 and Jun 2014. In both cases we use the micro-aethelometer AE-51 to measure the BC concentration and absorption coefficient with time resolution of 1 sec and flow speed of 0.2 l/min. The CHM15K ceilometer (1064 nm) and LM-10 lidar (532 nm) were used to obtain aerosol extinction coefficient profiles.

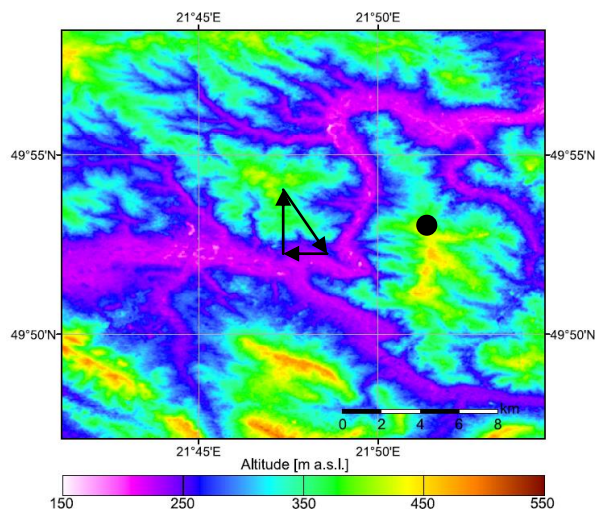


Fig.1 Topography of research area. The black circle shows the research station SolarAOT localized about 200 meters above valley of Strzyzow. The three arrows the indicate the hiking trail during micro-aethalometer measurements between bottom of valley and small hill 100 meter above.

The UAV for atmospheric research was built in 2013 by Synergy Technologies. Its wingspan is 2.4 m, length 1.8 m, while the take-off weight approx. 4.5 kg, and 0.7 kg load capacity approx. The aircraft has an autonomous control system that enables you to perform vertical profiles from the ground to a height of about 1.5 km. The UAV is equipped with a micro-aethalometer AE-51 and Vaisala radiosonde RS92SGP.

The main research station SolarAOT is equipped with aerosol in-situ instruments such as a seven wavelength aethalometer AE-31, three wavelengths polar nephelometer Aurora 4000, two photoacoustic extinctionometers (PAX) working at 532 and 870 nm respectively as well as the CIMEL 8N-EDPS9 (AERONET).

3. DATA PROCESSING

The data obtained from micro-aethelometer device includes nonrealistic negative values when sampling is performed at clear conditions or at a high time-resolution. In this case the instruments noise can cause the ATN values to remain unchanged or even to decline slightly between time steps. To reduce the signal to noise ratio filter methods are used. Hangle et al. (2011) developed ONA algorithm to eliminate the negative values. For this purpose the minimum delta attenuation (Δ ATN) value as assumed to defined the time frame of data averaging. However, we developed an alternative method which focus on the averaging of the ATN signal before the signal derivative. To do this we use the run mean filter with time frame of 20 sec. After reduction of ATN noise the aerosol absorbing coefficient and BC concentration are computed from ATN derivative

$$\sigma_{\text{ABS}} = \frac{d\text{ATN}}{dt} \frac{A}{Q \cdot C \cdot R(\text{ATN})} \quad (1)$$

$$\text{BC} = \frac{\sigma_{\text{ABS}} \cdot C \cdot R(\text{ATN})}{\sigma_{\text{ATN}}} \quad (2)$$

where, A is a sample spot area ($7.1 \cdot 10^{-6} \text{ m}^2$), Q is the volumetric flow rate ($2.5 \cdot 10^{-6} \text{ m}^3/\text{s}$), C is the multiple scattering optical enhancement factor (2.05 ± 0.03 Ferrero et al., 2011), R(ATN) is aerosol loading factor, and σ_{ATN} is the apparent mass attenuation cross section ($12.5 \text{ m}^2/\text{g}$ at 880 nm). The R(ATN) term compensates for the nonlinear (loading effect) due to increase of aerosol absorption over time, which in turn results in a reduction in the optical path. Schmid et al. 2006 found that it is needed only when ATN becomes higher than 20. Therefore we changed the filter when the ATN exceed this threshold value. Although this method may produce still negative values, however the improving of SNR is significant.

4. CASE STUDY

In this section we presents results of vertical profile measurements during winter season 2013/2014. We focused on the afternoon and night inversion condition in the lower PBL. Fig. 2 shows vertical profiles of BC concentration and absorbing coefficient in the first 100 m as well as

the single scattering properties at the SolarAOT station and weather conditions during measurements. Although relatively low inversion ($1^\circ/100\text{m}$) the BC concentration reaches the $60 \mu\text{g}/\text{m}^3$ (20 times larger than long-term mean for Dec) just above the valley bottom. 100 meters above the BC drops to about $5 \mu\text{g}/\text{m}^3$ and 200 m to $4.7 \mu\text{g}/\text{m}^3$ which is only two times larger than long-term mean). Similarly to BC concentration we found large vertical variability of aerosol absorption coefficient (Fig. 2 right panel). Extremely high value of absorption coefficient was measured close to valley bottom (about 0.8 km^{-1}). Integrating of absorbing coefficient in the first 200 m we estimated the absorbing aerosol optical depth (AAOD) of 0.044 (at 500 nm). In addition, based on the single scattering albedo (SSA) observation at SolarAOT station (0.86) and vertical profiles of absorbing coefficient we computed the AOD in the first 200 m. The AOD was about 0.31 (500 nm). This value cannot be

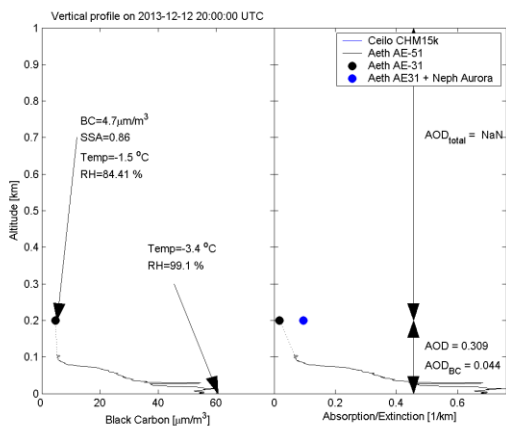


Fig. 2 Vertical profiles of BC concentration [$\mu\text{g}/\text{m}^3$] (left panel) and absorption coefficient [km^{-1}] (right panel) at 20 UTC on Dec 12, 2013.

compare to the total AOD measured at CIMEL because of the cloud condition which was observed during day. Aerosol instantaneous RF obtained from the MODTRAN code for shortwave range shows negative value close to surface (about $-10 \text{ W}/\text{m}^2$) and strong positive in the upper levels (Fig 3 left). The largest aerosol impact of radiation balance was found during high solar zenith angle (SZA) due to large air mass and large absorption of solar radiation in the atmosphere. As results of high BC concentration the radiative

heating shows maximum close to surface (Fig. 3 right). In case of SZA equal 70° the radiative heating exceeds $20 \text{ K}/\text{day}$ and for 80° reaches $60 \text{ K}/\text{day}$. At the altitude of SolarAOT station the radiative heating is between 5 and $10 \text{ K}/\text{day}$.

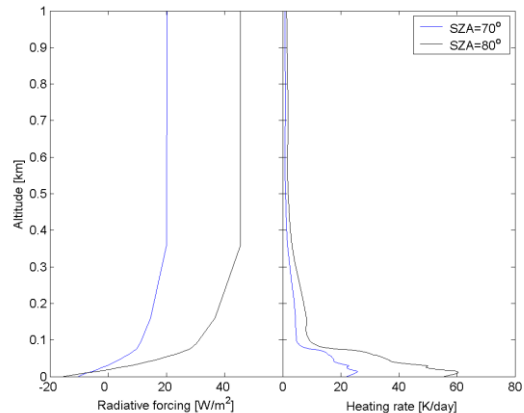


Fig. 3 Vertical profiles of aerosol RF [W/m^2] (right panel) and heating rate [K/day] (left panel) for solar zenith angle 70° (blue line) and 80° (black line) obtained from MODTRAN simulation performed for shortwave spectrum and for aerosol optical properties measured on Dec 12, 2013.

Similar strong vertical variability of BC concentration and aerosol optical properties was observed on Feb 3, 2014. In this case weather condition was extremely different between SolarAOT station and bottom of valley. For example temperature and relative humidity in the upper station was $+4.6^\circ$ and 28.1% while in the lower -6.9° and 92.2% respectively. In addition, the CIMEL observation during day shows very clear air mass with AOD at 500 nm only 0.039. During night (20 UTC) the BC concentration exceed $35 \mu\text{g}/\text{m}^3$ and aerosol absorbing coefficient 0.45 km^{-1} at the valley bottom. Estimated the AAOD in the first 200 m was 0.019 and AOD was 0.181. SSA at the SolarAOT station was 0.89 indicating moderate absorbing particles. Simulation of RF shows $-10 \text{ W}/\text{m}^2$ at the surface, almost zero several meter above the valley surface and increasing with altitude positive RF above. Radiative heating shows strong variability in the first 100 m from $35 \text{ K}/\text{day}$ at the surface to $7 \text{ K}/\text{day}$ 100 m above for SZA of 80° . Both examples shows very strong variability of BC, aerosol, and radiative properties of with altitude as a results of the very weak vertical mixing and local emission of air pollution during heating season. Very high value of BC leads to large radiative heating during day which intensively the inversion.

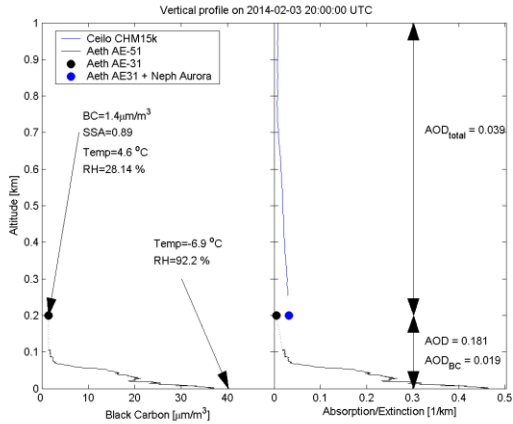


Fig. 4 Vertical profiles of BC concentration [$\mu\text{g}/\text{m}^3$] (left panel) and absorption coefficient [km^{-1}] (black line) as well as extinction coefficient from ceilometer (blue line of right panel) at 20 UTC on Feb 3, 2014.

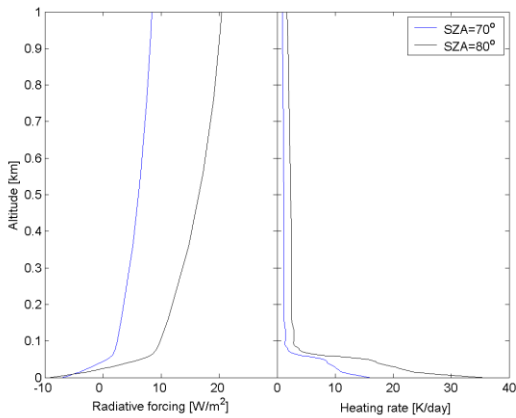


Fig. 5 The same as the Fig 3 but at 20 UTC on Feb 3, 2014. In this case the extinction coefficient come from the ceilometer observation at 1064 nm.

5. FLIGHT EXPERIMENTS

In this section we present results of flight measurements performed in Sep 2013 and in Jun 2014 at SolarAOT station in Strzyzow. Fig. 6 shows profiles of aerosol extinction coefficient at 532 nm obtained from LB10 lidar (blue line), absorption coefficient (black line) measured by micro-aethalometer (a) and BC concentration in $\mu\text{g}/\text{m}^3$ (b). The blue dots and black squares at the surface (altitude of SolarAOT station) corresponds to ground-based observation made by aethalometer AE-31, Aurora 4000, and photoacoustic extinctionionometer (PAX)

respectively. In case on Lidar the extinction coefficient below the 500 m (overlap altitude) is linearly extrapolated to the surface value measured by aethalometer and nephelometer. Data from micro-aethalometer shows relatively small variation with altitude, however, the noise is high especially close to surface. The high noise is a results of relatively clear air mass. The AOD was 0.07 at 500 nm, surface BC about $0.4 \mu\text{g}/\text{m}^3$ and columnar SSA 0.89 and surface SSA 0.91. Also, the micro-aethalometer tends to overestimated the BC concentration and absorption coefficient. Small variation with altitude can be explain by vertical mixing. Data from radiosonde shows almost constant potential temperature up to 1000 m.

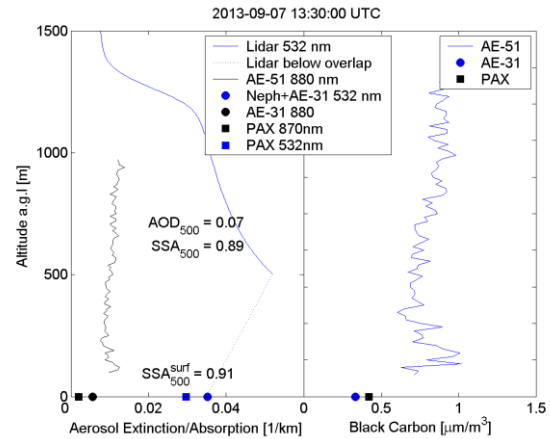


Fig. 6 Profiles of aerosol extinction estimated from LB-10 lidar at 532 nm as well as the absorption coefficient (a) and BC concentration (b) measured by micro-aethalometer onboard the UAV at 13 UTC on Sep 7, 2013.

Fig. 7 presents results from flight performed on Jun 10, 2014 during morning condition. In this case both micro-aethalometer and ceilometer data show multilayers structure of the PBL. The flight measurements have better SNR due to more polluted conditions. During this day the AOD was 0.195 (500 nm), columnar SSA of 0.95 and surface SSA of 0.91, while surface BC was $1.7 \mu\text{g}/\text{m}^3$.

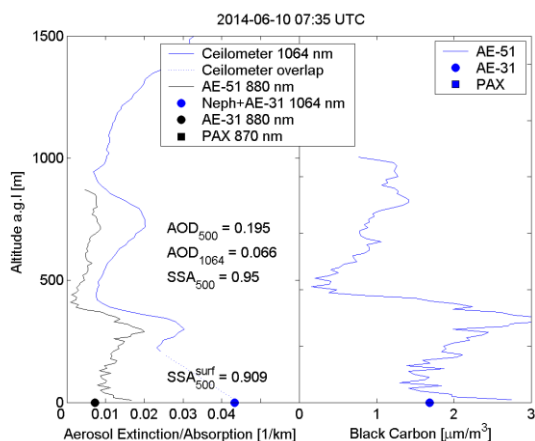


Fig. 7 The same as Fig. 6 but at 07:35 UTC on Jun 10, 2014.

6. CONCLUSION

We found large variability of BC concentration and absorbing coefficient with altitude during inversion conditions. The haze layers have usually less than 100 m of depth with maximum of BC concentration at the bottom of valley or several meters above. Estimated BC optical depth in the first 200 meters was up to 0.04 (at 500 nm) which corresponds to very high (up to 60 K/day) radiative heating during day and strong negative RF at the surface and positive RF several meters above.

The UAV test flights show that maximum flow rate (0.2 l/min) in the micro-aethalometer AE-51 is too low to get sufficient SNR during clear air mass condition. In this case measurements with vertical speed about 3-4 m/s required the flow rate between 0.5-0.7 l/min. We have plan to increase it by using different pump. The second limit is related to lidar/ceilometer overlap problem. Due to this the vertical profiles of extinction coefficient can be used above the 200-400 m a.g.l. Therefore we are developing a near field optics in the our new Polly^{XT} lidar to retrieve aerosol optical properties from 50 m above the device. In this case the synergy with micro-aethalometer UAV observation allows to retrieve the profiles of SSA between 50 and 1000 m.

7. ACKNOWLEDGMENTS

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