SOURCE APPORTIONMENT AND OPTICAL PROPERTIES OF BLACK CARBON AEROSOL PARTICLES

FIZINIŲ IR TECHNOLOGIJOS MOKSLŲ CENTRAS

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Introduction

In recent years, the aerosol impact on the Earth's radiative balance had been widely recognized. The influence of aerosol particles on climate and ecosystems comes from the ability to scatter or absorb solar radiation and alter optical properties of clouds. Black carbon (BC) is the most strongly **light-absorbing** component of aerosol particles, which is formed by the incomplete combustion, i.e. anthropogenic (fossil fuels, biofuels, and biomass) and natural (wildfires, volcanoes) and can stay in the atmosphere for up to 2 weeks.

BC exhibits noticeably strong radiative forcing alike greenhouse gases. Its significance as the **second highest** anthropogenic contributor for global warming after CO_2 was acknowledged in special report *Global Warming of 1.5 ºC* by IPCC (Masson-Delmotte et al.). And yet, the net impact of BC is still unknown as it can influence climate through multiple mechanisms (Schacht et al.). Consequently, understanding the behavior and source apportionment of BC has a significant impact on global and regional climate change.

Emissions of fine and coarse aerosol particles are increasing due to growing use of biomass burning, fossil fuel combustion, exhaust emissions and energy extraction in power plants. Considering that in the European Union (EU) at present there are two threshold values set for PM_{10} concentration, more measures towards controlling pollutant emissions have to be taken, especially for emissions of BC which is considered as **short-lived climate forcer**.

The aim of study was to perform a systematic source apportionment of BC by examining dynamics of equivalent mass concentration and optical properties during heating season. Furthermore, the research aims to select the most suitable values of AAE for biomass burning and fossil fuel combustion source apportionment.

Methods

In our study a 7-wavelength Aethalometer (Magee Scientific, EA31) was deployed in Vilnius (urban background site) in October 2014 – April 2015.

The Aethalometer model for BC source apportionment was chosen as it only requires a multi-wavelength light absorption dataset and can attribute BC to two general source categories: biomass burning and fossil fuel combustion (Zotter et al.). The method is based on the dependence of aerosol absorption on the wavelength of light which is defined as source specific **absorption Ångström exponent (AAE)**. AAE describes the wavelength variation in aerosol absorption. The relation between AAE and b_{abs} is defined as:

$$b_{\mathrm{abs}}(\lambda) = b_0 \lambda^{-\mathrm{AAE}},$$

where λ - wavelength, b_{abs} - aerosol absorption coefficient and b_0 wavelength independent constant. In order to separate BC_{ff} (BC originated from fossil fuel combustion) and BC_{bb} (BC originated from biomass burning) the most suitable values of AAE (absorption Ångström exponent) were selected (0.90 and 2.09), respectively.

Levoglucosan is known as a main marker of wood burning. In order to observe its appearance, an Aerosol Chemical Speciation Monitor (ACSM) was used and full organic aerosol mass spectra was measured. Intensity of the signal *m/z* 60 was normalised to organic fraction (f60) and was used as a proxy of levoglucosan.

Results

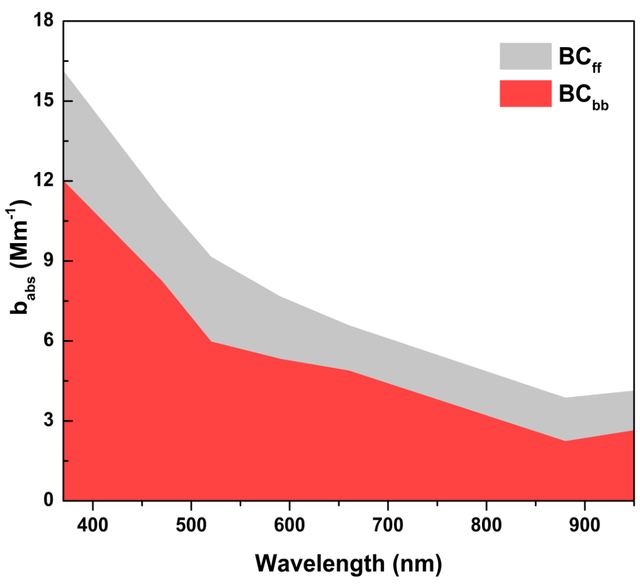

Figure 1 Absorption coefficients of BC_{ff}, BC_{bb} as a function of wavelength in range 370-950 nm.

Present results show that BC originating from biomass burning is responsible for the strongest absorption in the whole measured spectrum (370-950 nm), exhibiting the highest absorption coefficient values at shorter wavelengths (370-470 nm) (Figure 1). Absorption of BC_{bb} is 2.4 times stronger comparing to BC_{ff} thorough the spectrum.

The contrast in absorption coefficient values of BC_{bb} and BC_{ff} comes from the differences in chemical composition. Aerosol pariticles originating from incomplete biomass burning contains BC_{bb} and higher CO/NO_X ratio than BC_{ff}. Fossil fuel combustion in motor vehicle engines takes place at relatively higher temperatures, thus the originating aerosol particles contain BC_{ff} and higher amount of NO_X and exhibit weak absorption in near-UV.

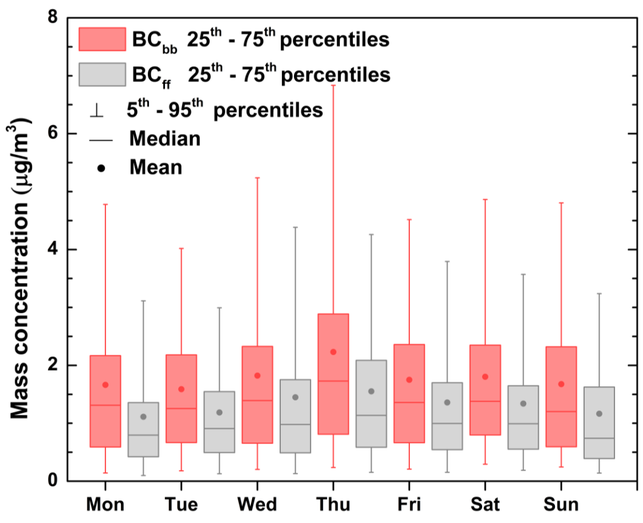

Figure 2 The dynamics of BC_{bb} and BC_{ff} mass concentrations daily over the course of the week.

Over the course of the week (Figure 2) the 25th percentile values varies in ranges 0.6-0.8 µg/m³ and 0.4-0.6 µg/m³ for BC_{bb} and BC_{ff}, respectively. However, higher oscillations are observed for the values of 75th percentile: 2.2-2.9 µg/m³ and 1.4-2.1 µg/m³ for BC_{bb} and BC_{ff}, respectively. Mean daily mass concentrations of BC_{bb} are 1.4 higher than BC_{ff}. This can be explained due to more active biomass burning for residential heating. Highest BC_{bb} and BC_{ff} mass concentration values are observed on Thursdays. The oscillations observed over the course of the week are related with transport use which is more active during workweek and passive on weekends.

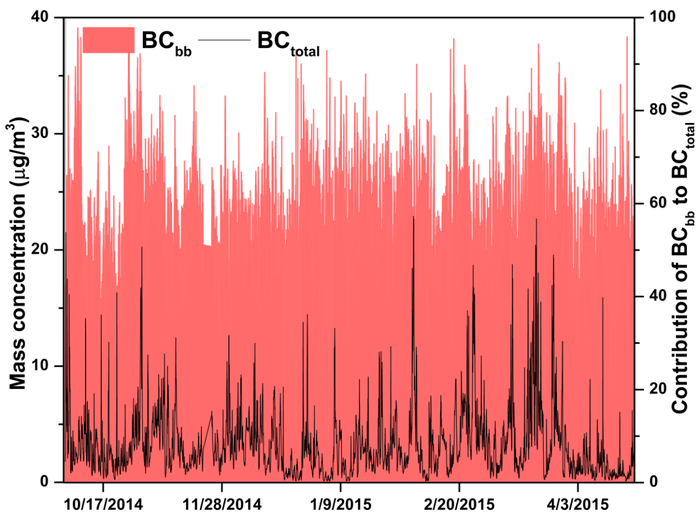

Figure 3 Time series of total BC mass concentration and contribution of BC_{bb} to total BC.

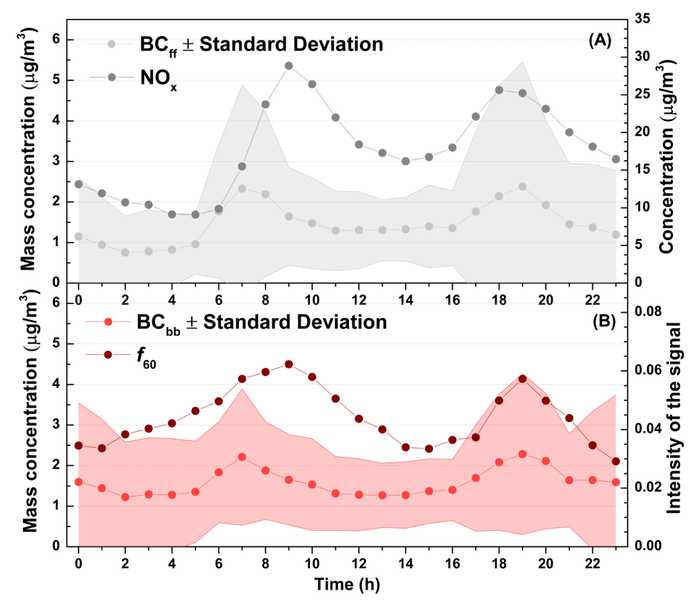

Figure 4 Diurnal cycles of BC_{ff}, NO_X (A), BC_{bb} and f_{60} (B).

The mean BC mass concentration during the measurement campaign was 3.00 µg/m³ (Figure 3). Moreover, our results indicate that BC originating from biomass burning accounts for a major part of total BC (59.3 %).

Diurnal cycles of BC_{ff} and BC_{bb} mass concentrations show strong variations over the course of the day (Figure 4). Both diurnal cycles exhibit peaks corresponding with morning (6-8AM) and evening (5-8PM) rush hours. However, BC_{ff} mass concentration values with standard deviation (SD) show stronger variation during the rush hours in comparison to BC_{bb}. BC_{ff} contributes to total BC mass concentration 49.1 % during daytime (10AM-4PM) and 41.2 % during nighttime (9PM-5AM). Thus, indicating active transport use during the daytime and decrease in emissions from traffic at nighttime.

Diurnal cycle of NO_X hourly mean concentration exhibits good correlation with BC_{ff} (r=0.66). Therefore, it can be concluded that AAE_{ff} value 0.9 is suitable for apportioning BC_{ff} from total BC.

In contrast to BC_{ff}, BC_{bb} mass concentration ± SD values show stronger variation at nighttime indicating an increase in the number of individual houses being heated by biomass burning.

In order to support validity of AAE_{bb} value selection, diurnal cycle of f_{60} is graphed together with diurnal cycle of BC_{bb}. Good correlation was found between BC_{bb} and f_{60} (r=0.83) thus indicating that AAE_{bb} value 2.09 is suitable for separating BC_{bb} from total BC during heating season in urban environment. Good correspondence of Aethalometer model source apportionment results with biomass burning related organic aerosol parameters suggests that source apportionment was performed successfully.

Table 1 Statistical analysis of BC mass concentration dynamics over the course of a week (µg/m³)

Weekday	BC source	Mean	Standard deviation	Median	Maximum	25th percentile	75th percentile
Monday	BCbb	1.7	1.5	1.3	14.1	0.6	2.2
	BCff	1.1	1.3	0.8	25.6	0.4	1.4
Tuesday	BCbb	1.6	1.3	1.3	14.5	0.7	2.2
	BCff	1.2	1.1	0.9	14.5	0.5	1.5
Wednesday	BCbb	1.8	1.7	1.4	14.2	0.7	2.3
	BCff	1.4	1.7	1.0	22.0	0.5	1.8
Thursday	BCbb	2.2	2.1	1.7	14.4	0.8	2.9
	BCff	1.6	1.4	1.1	19.5	0.6	2.1
Friday	BCbb	1.8	1.6	1.4	14.9	0.7	2.4
	BCff	1.4	1.4	1.0	17.7	0.5	1.7
Saturday	BCbb	1.8	1.5	1.4	15.4	0.8	2.3
	BCff	1.3	1.3	1.0	15.8	0.6	1.6
Sunday	BCbb	1.7	1.5	1.2	11.0	0.6	2.3
	BCff	1.2	1.2	0.7	16.7	0.4	1.6

Conclusions

Our results indicate that BC originating from biomass burning contributes a major part (**59.3 %**) to total BC mass concentration during heating season. Although the dynamics of total BC mass concentrations exhibit strong variations over the course of the day and over the course of the week due to strong increase in traffic exhaust emissions. The most suitable AAE_{bb} AAE_{ff} values for BC_{bb} and BC_{ff} source apportionment were chosen to be 2.09 and 0.9, respectively.

References

- Masson-Delmotte *et al.*, "Global Warming of 1.5 °C. An IPCC special report on the impacts of global warming of 1.5 °C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change" 2018.
- Schacht *et al.*, "The importance of the representation of air pollution emissions for the modeled distribution and radiative effects of black carbon in the Arctic," *Atmos. Chem. Phys. Discuss.*, pp. 1–39, 2019.
- Zotter, Peter, et al. "Evaluation of the Absorption Ångström Exponents for Traffic and Wood Burning in the Aethalometer-Based Source Apportionment Using Radiocarbon Measurements of Ambient Aerosol." *Atmospheric Chemistry and Physics*, vol. 17, no. 6, pp. 4229–49, 2017.

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