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Modelling of black carbon statistical distribution and return periods of extreme concentrations☆

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ABSTRACT

Eight datasets of 1-h black carbon (BC) concentrations measured in Warsaw agglomeration (Poland), at urban background and sub-urban sites, and in Racibórz, a small town in Upper Silesia district (regional background site) were analyzed to evaluate BC levels, daily profiles and statistical distributions of concentrations in Central-Eastern European region. The observed mean levels ranged from 1483 ng m^{-3} in suburban site during summer to 3358 ng m^{-3} in regional background site in winter. Observed diurnal patterns were bimodal in the locations dominated by traffic emissions, but unimodal, with elevated evening peak in individually heated residential area. Three theoretical frequency distributions were applied to fit analyzed datasets separately. The lognormal distribution was the most appropriate to represent the middle-range values, while the high concentrations were satisfactorily predicted by the type I two-parameter exponential distribution which was used to estimate the return periods of extreme concentrations for winter months.

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1. Introduction

Atmospheric aerosols play fundamental role in the functioning of the Earth system, and their load have been included as one out of nine major processes within the framework of planetary boundaries which define a safe operating space for humanity, based on the intrinsic biophysical processes which regulate the stability of the Earth system (Rockström et al., 2009; Steffen et al., 2015). Atmospheric aerosols have well-known, serious human health impacts, leading to about 3.7 million deaths per year (WHO, 2014), as well as a broad spectrum of effects on the climate of the Earth and functioning of the planet's system. One of the most important components of the atmospheric aerosol is carbonaceous matter of natural and anthropogenic origin, which can be primary or secondary and contain both inorganic and organic carbon (OC) species. Due to its distinct properties and extensive range of effects, atmospheric carbonaceous matter – and black carbon (BC) in particular – is a subject of various research fields, such as climate change, air chemistry, ambient air quality, biogeochemistry, as well as paleoclimatology. Among such a wide group of researchers, it has been difficult to agree on a clear, unambiguous terminology concerning quantification of carbonaceous matter. Therefore, Petzold et al. (2013) have lately proposed a consistent terminology, based on the carbonaceous material properties and associated measurement techniques and instruments. According to this recommendation, 'soot' is a qualitative term tightly related to the source of emission of carbonaceous matter, and denotes carbonaceous particles formed from incomplete combustion of carbon-based fuels, i.e. fossil fuels, as well as biofuels and biomass. Two main measurement techniques are used to determine the ambient concentrations of inorganic carbonaceous pollutants, and these are the filter-based light absorption (optical) method whose results are referred to as BC, and the thermal-optical method which provides measurements expressed as elemental carbon (EC).

According to the strict definition, BC is the ideally light-absorbing pollutant, refractory and insoluble in water and organic solvents, which is composed of aggregates of small carbon spherules that consist mostly of graphite-like sp2-bonded carbon atoms (Petzold et al., 2013). However, such definition is not useful in practice, as carbonaceous matter in the atmosphere is never a pure matter, but a mixture of various carbonaceous compounds of

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different properties. In particular, there are other than BC light-absorbing species, such as brown carbon (BrC), which may influence the BC measurement results based on the optical method. Correction of such bias is possible, but requires a multi-wavelength instrument. In present study, BC concentrations were determined via absorbance measurement by a single-wavelength (880 nm) aethalometer, further converted into concentration units with the use of calibration factor provided by the manufacturer.

BC particles are contained in the fine aerosol mode, therefore they are often transported over long distances, mixing with other aerosols along the way. The aerosol mix can form transcontinental plumes of atmospheric brown clouds, with vertical extents of 3–5 km (Ramanathan and Carmichael, 2008). Over time, BC aerosol ages and becomes coated by water and secondary species, including sulfate, ammonium, nitrate and organics. Such coating significantly changes the optical properties of particles (Moffet and Prather, 2009).

According to a broad review by Putaud et al. (2010), in Central Europe BC constitutes on average 5% of rural background $PM_{2.5}$ (Particulate Matter with aerodynamic diameter less than 2.5 μm), 14% of urban background $PM_{2.5}$ and 21% of $PM_{2.5}$ collected in kerbside sites. The fraction of <150 nm diameter (which contains over 80% of particle number in the atmosphere), consists almost entirely of organic and inorganic carbon aerosol (Putaud et al., 2010). In Central-Eastern Europe, the share of EC in $PM_{2.5}$ seems to be higher. The 2010 measurement campaign conducted in Poland has shown that EC constitutes on average 14% of $PM_{2.5}$ mass in the EMEP rural background monitoring site in Diabla Góra (Northern-Eastern Poland), and 21% in the urban background site in Katowice in Upper Silesia region of Southern Poland (see Fig. 1). The latter number increases to 25%, when only winter (DJF) measurements are taken into account (GIOŚ, 2011).

In general, atmospheric aerosols play an important role in the global climate system through modifications of the global radiation budget (Stier et al., 2007): (1) directly, by scattering and absorption of radiation; (2) indirectly, by the modification of cloud properties and abundance; and (3) semi-directly, by the effect of direct and indirect aerosol effects on cloud properties and abundance via modification of the thermal structure of the atmosphere and the surface energy budget. Due to its strong light-absorbing potential, BC modifies the optical properties of the aerosol. However, especially in areas affected by severe air pollution originating from combustion processes, the usual effect of the Earth's surface dimming caused by atmospheric aerosol (negative Radiative Forcing) (IPCC, 2013) can be reduced, or even reversed, by the absorption of solar energy by BC, which can lead to an increased greenhouse effect (positive Radiative Forcing). Recent investigations suggest that BC – after CO_2 – is the second largest global warming contributor (Bond et al., 2013), as per unit of mass in the atmosphere, it can absorb a million times more energy than CO_2 (Bond and Sun, 2005). Simultaneously, the frequently emphasized negative impact of aerosols on human health gets continuously reinforced proofs of being associated only with its selected components and fractions, especially due to the carbonaceous particles. It has recently been confirmed, that at least short-term health effect associations with BC are more robust than those with $PM_{2.5}$ or PM_{10} (WHO, 2013). Meta-analysis of 34 studies conducted by Janssen et al. (2011) demonstrated that the increase of BS (black smoke, light-absorbing particles indicated by the reflectometric method, applied in Europe since the 1920's, nowadays displaced by the gravimetric measurements of PM) concentration by 10 $\mu g\ m^{-3}$ leads to the increase of mortality rates by 0.9% and 0.95% in case of the cardiovascular and respiratory causes, respectively. Toxicological studies suggest that BC may not be directly toxic, but it may operate as a universal carrier of a wide variety of combustion-derived chemical constituents of varying toxicity to sensitive targets in the human body such as the lungs, the body's immune system cells and possibly the systemic blood circulation (WHO, 2012). Finally, the wide environmental impacts of BC are due to its ubiquitous occurrence in atmospheric air, soils and marine sediments. Key factor which determines aerosol impacts on plants is the rate of deposition which is determined both by the characteristics of particles (e.g. their size) and the roughness of leaves. The deposition velocity has been shown to be higher for ultrafine particles than for the coarse ones, resulting in deposition of 0.5–5 μg of $PM_{0.2}$ per 1 cm^2 of leaf surface, with *Betula pendula* – silver birch – being the most efficient particle collector amongst the most popular tree species in Poland (Sæbø et al., 2012).

Virtually all thermal processes involving combustion of matter rich in carbon are the emission sources of carbonaceous aerosol in ambient air. These are mainly fuel combustion (fossil fuels and biofuels) and open biomass burning (forest fires, savanna/grass

Fig. 1. Location of the BC measurement sites in Poland. Sites investigated by this study are marked with dots, and the sites for which BC data were reported by other Authors – with triangles.

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