A seven-years based characterisation of aerosol light scattering properties at Central European rural site: Variability and Source apportionment

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Introduction

Atmospheric aerosols have a significant impact on the radiative forcing of Earth's climate, either directly through aerosol-radiative interactions (ARIs, scattering or absorption of radiation) or indirectly through aerosol-cloud interactions (ACIs). The aerosol direct effect could promote or suppress positive forcing [2,3,4,5,7]. The predominant cooling effect results from the scattering of radiation by certain types of atmospheric aerosols which reduces the amount of solar radiation reaching the Earth's surface[4]. This phenomenon offsets the greenhouse effect and alters the radiation balance[6]. Radiative forcing caused by aerosols remains one of the main sources of uncertainty in climate modelling.

Objective

The aim of this study is to focus on the temporal variations of light-scattering aerosols at a rural site in Central Europe. The total light scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients and associated calculated optical properties such as the Ångström exponent (*SAE*), backscattering ratio (*b*), and asymmetry factor (*g*) are characterized considering different time scales (annual, seasonal, monthly, or diurnal) based on long-term measurement. The optical properties are compared for days with and without fog event and different categories of sky cloudiness, with particle number size distribution, potential sources of atmospheric aerosols and the influence of meteorological conditions and other atmospheric pollutants at the National Atmospheric Observatory Košetice (NAOK) to better understand the direct effects of aerosols on the local climate.

Assymetry factor represents the probability that radiation is scattered in a given direction (-1 for complete backscattering, 1 for complete forward scattering) and is based on *b*:

$$g = -7.143889b^3 + 7.464439b^2 - 3.9356b + 0.9893$$

Methods

Measurements were carried out continuously from August 2012 to December 2019 at the rural background site National Atmospheric Observatory Košetice (NAOK; 49°34'20.787 "N, 15°4'48.155 "E) located in the central part of the Czech Republic (Fig. 1).

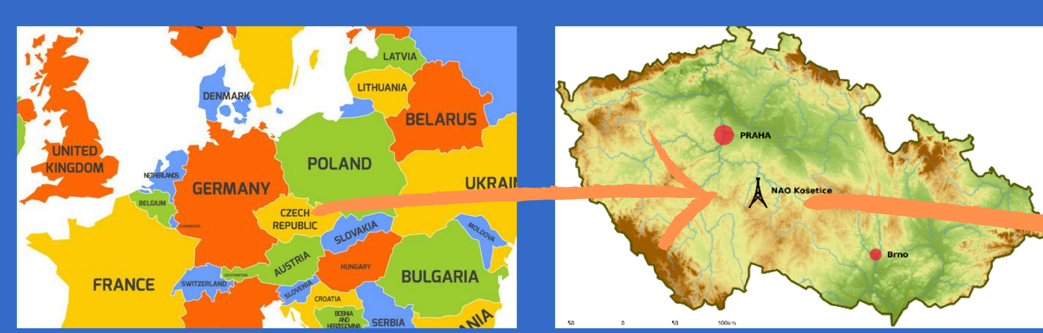


Fig. 1 Location of National Atmospheric Observatory Košetice

σ_{sp} (total scattering coefficient) and σ_{bsp} (backscattering coefficient) at 700, 550 and 450 nm were obtained from the Integrating Nephelometer TSI 3563 (Fig. 2) sampling at 4 m above the ground (RH<40%; PM10 sampling head - Leckel GmbH) with 5 min resolution. Correction for standard conditions, truncation and non/linear illumination was applied [1].

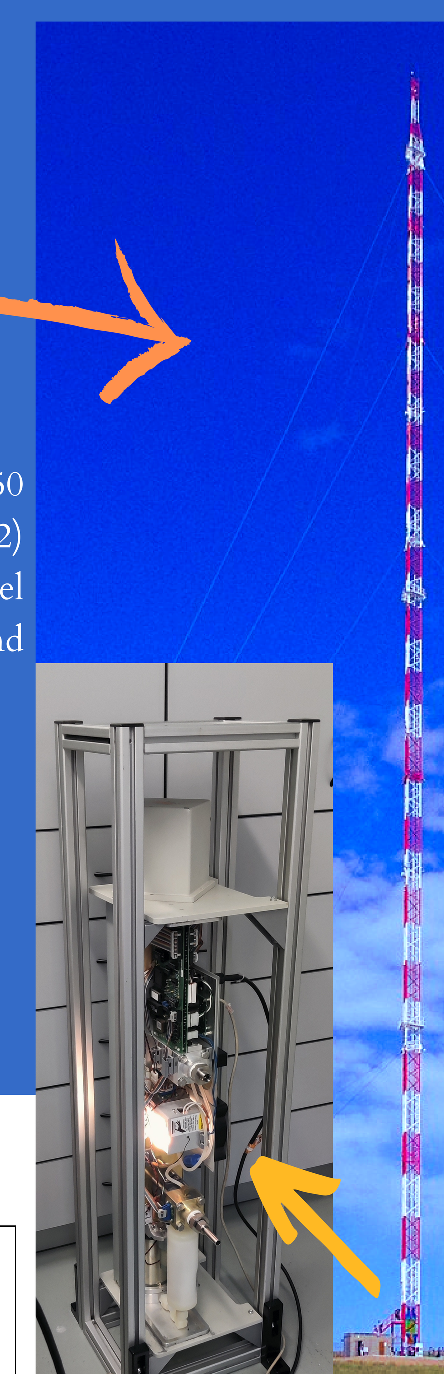


Fig. 2 Nephelometer TSI 3563 and atmospheric tall tower Košetice

Angström exponent expresses the dependence of particle size on the wavelength of scattered light (γ1 and γ2) through σ_{sp}:

$$SAE = \frac{\log(\sigma_{sp}(\gamma 1)) - \log(\sigma_{sp}(\gamma 2))}{\log(\gamma 1) - \log(\gamma 2)}$$

Backscattering ratio is defined as a ratio of backscattering and total scattering of light by aerosols at the same wavelength:

$$b = \frac{\sigma_{bsp}}{\sigma_{sp}}$$

Results

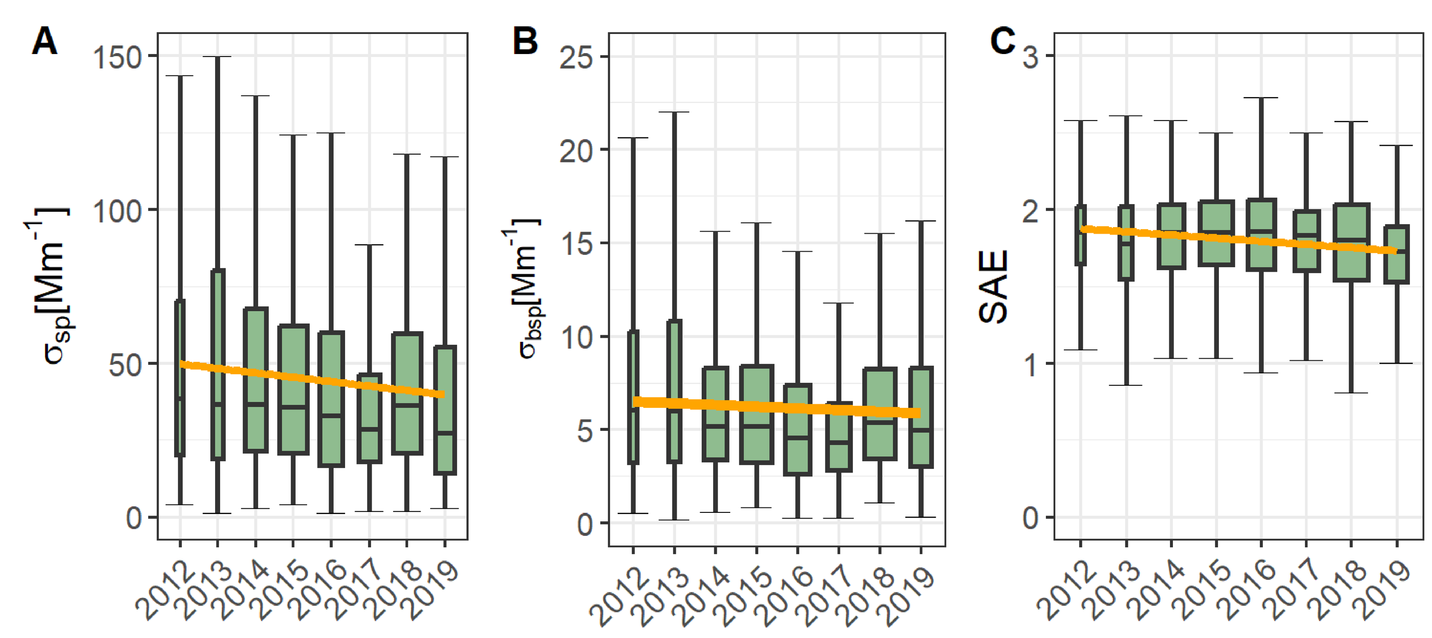


Fig. 3 Annual trend of σ_{sp}, σ_{bsp} and SAE at 550 nm. The black lines correspond to the median, green area denote to 25 and 75th percentile. The boxes are drawn with widths proportional to the square roots of "number of observations in separate years 2012-2019. The orange line represents trend which was significant (α=0.05, p<0.05).

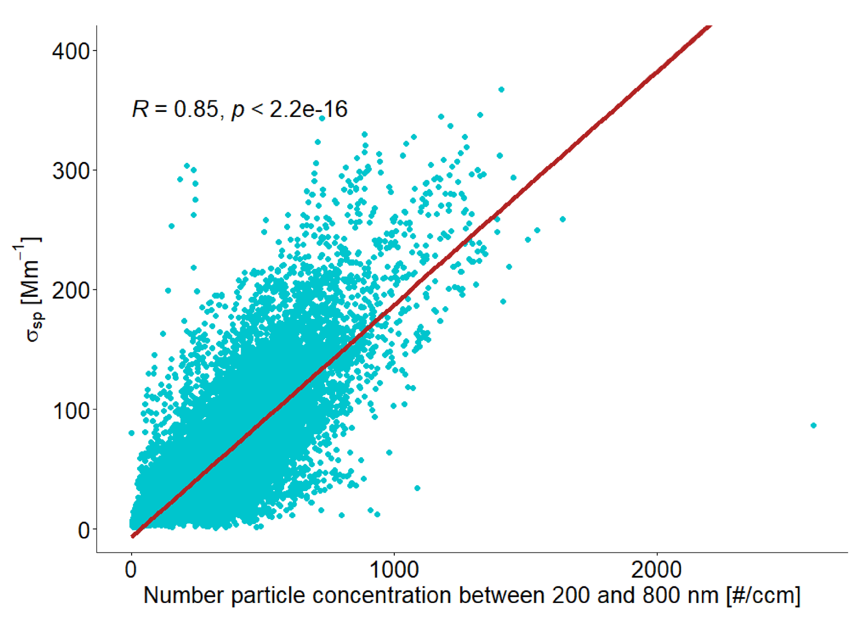


Fig. 4 The correlation plot with Spearman correlation coefficient R between σ_{sp} at 550 nm and the number concentration of particles between 200 and 800 nm.

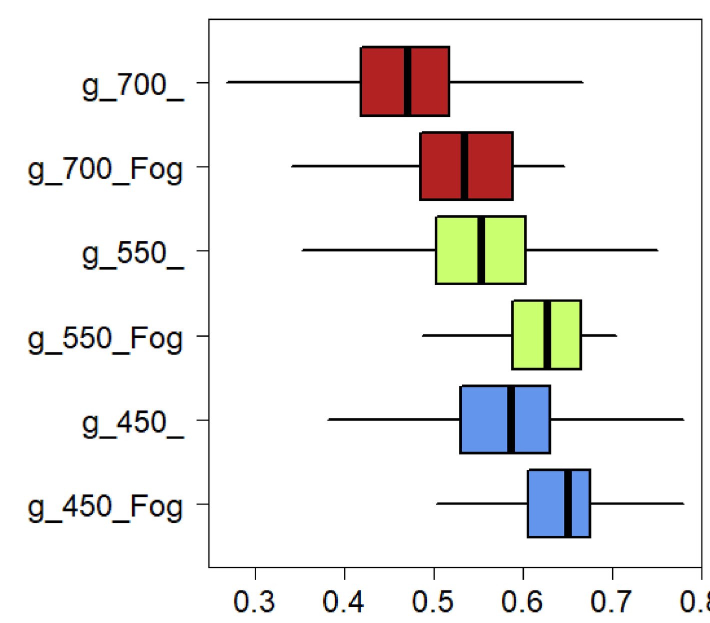
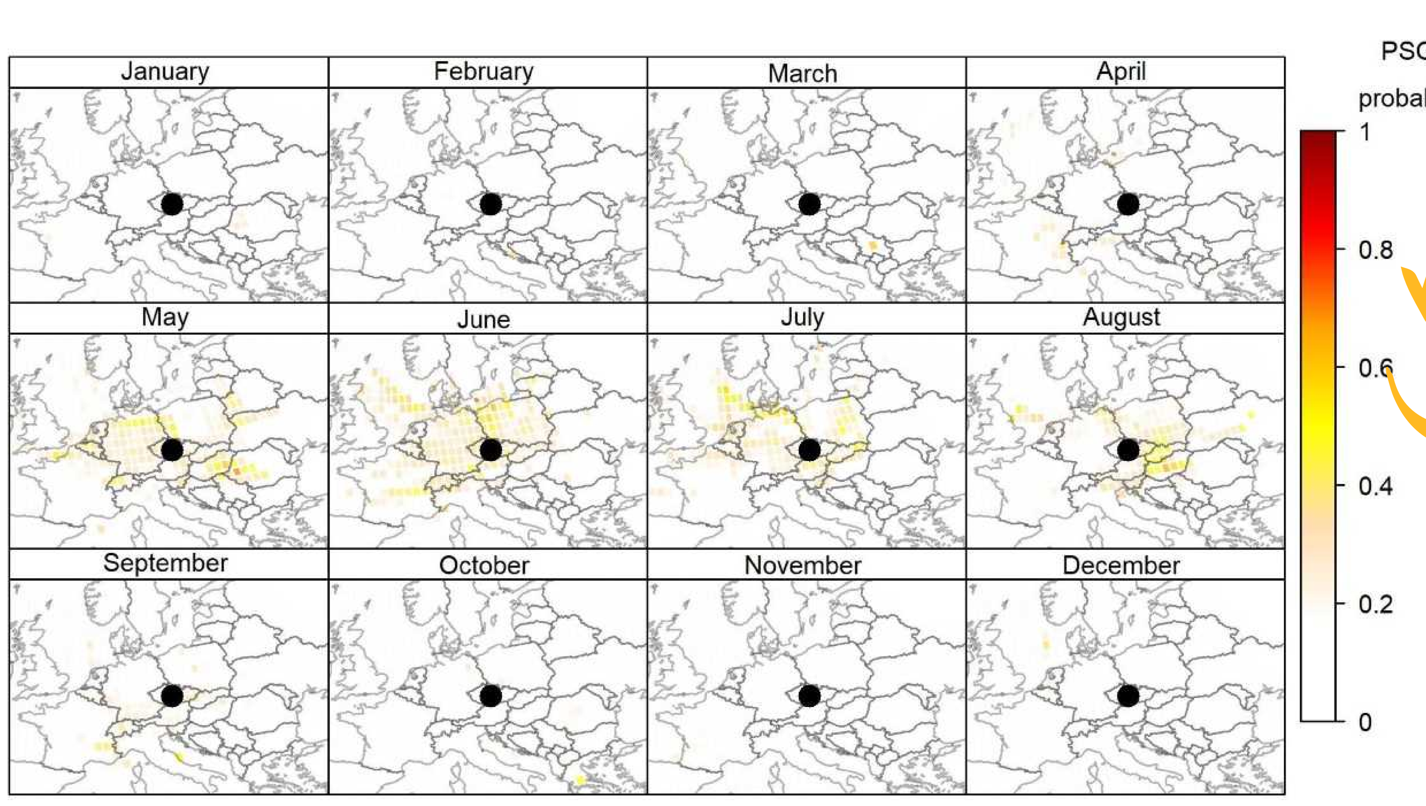


Fig. 5 Comparison of g during days with (Fog) and without (_) fog event at 450 nm (blue), 550 nm (green), and 700 nm (red) wavelength. The black line represents median, coloured areas 25th and 75th interval, empty circles outliers.



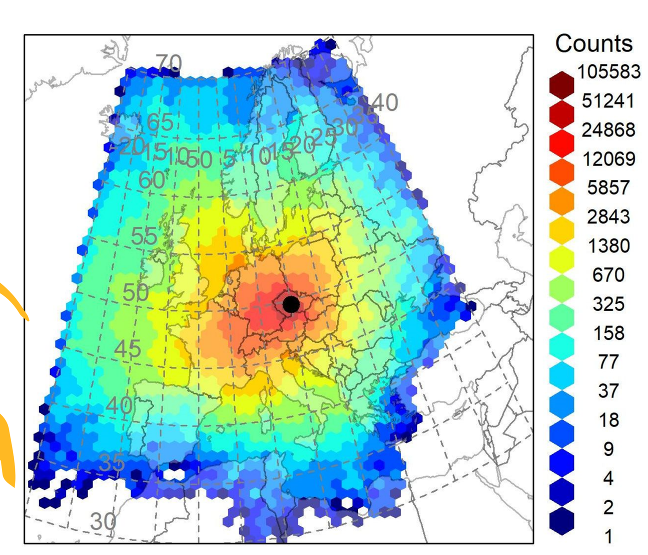


Fig. 6 PSCF of the SAE at 550 nm calculated separately for different months with monthly 75th percentile as the limit value. The location of NAOK is denoted with black circle. The colourful map shows the frequency of trajectories crosses specific grid cells.

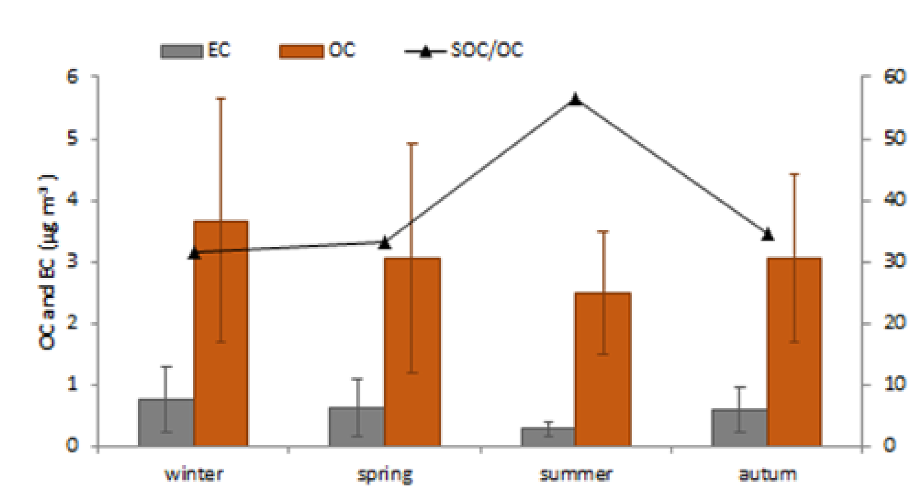


Fig.8 Seasonal variation of OC, EC and SOC/OC measured at NAOK (2013-2019).

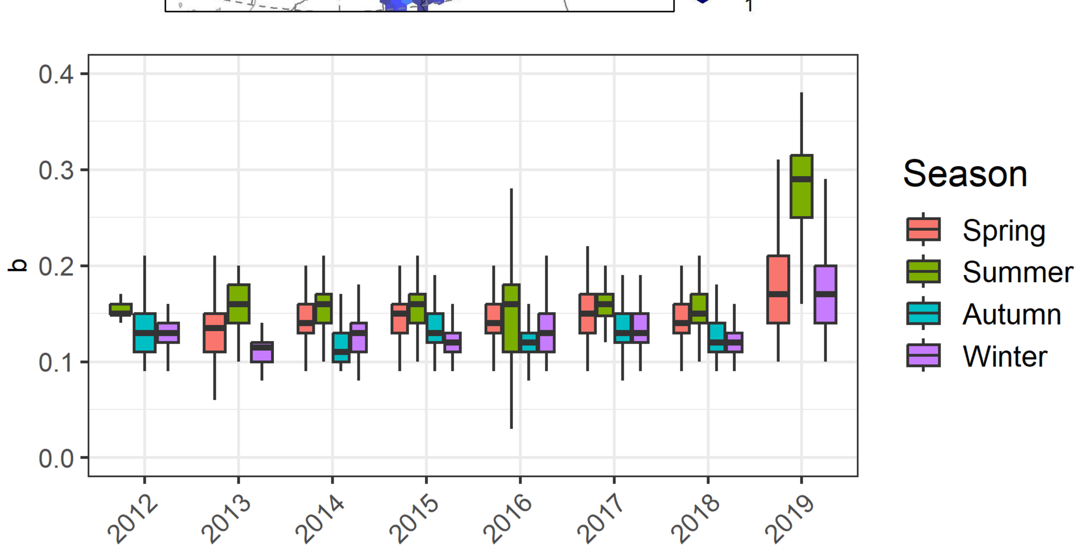


Fig. 7 Seasonal variation of b at 550 nm in individual years. Black line represents median, coloured area denote to 25 and 75th percentile.

Tab. 1 Annual median and slopes [unit/year] of median trends during 2012-2019 at NAOK

	450 nm		550 nm		700 nm	
	Median	Slope	Median	Slope	Median	Slope
σ_{sp}[Mm^{-1}]	51.2	-3.22	35.2	-2.50	22.5	-1.03
σ_{bsp} [Mm^{-1}]	6.5	-0.18	5.1	-0.18	4.3	-0.09
SAE	1.81	-0.002	1.82	-0.010	1.83	-0.018
b	0.12	0.015	0.14	0.012	0.18	0.013
g	0.60	-0.027	0.57	-0.021	0.48	-0.022

Summary and conclusions

- Overall decreasing annual trend of σ_{sp} and σ_{bsp} from 2012 to 2019
- Decreasing trend of *SAE* - shift to relative bigger particles with increasing *b* and decreasing *g*
- Increased efficiency of backscattering at lower concentration and bigger size
- Elevated *SAE* in summer - SOA formation vs decreased *SAE* in winter - aerosol aging + atm. stability
- Decreased *b* in winter - less backscattering species in winter (increase of carbonaceous aerosols)
- *g* higher during fog events - decreased backscattering compared to non-fog events = "multiple scattering"
- PSCF: an increased probability of source location identification of observed values over 75th percentile of SAE during late spring and summer months - mainly in NW, N and SE regions
- As the probability is not dense in any region, the results may be influenced by synoptic situation over Europe more than potential sources of aerosols
- The optical aerosol optical properties were correlated the most with the presence of particles between 200 - 800 nm
- The seasonal variation of scattering correlates to the seasonal variation of secondary organic aerosol formation

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This work was supported by the Ministry of Education, Youth and Sports of CR within the CzeCOS program, grant number LM2018123 within the National Sustainability Program I (NPU I), grant number LO1415, European Regional Development Fund-Project ACTRIS-CZ LM201818122, ACTRIS IMP (www.actris.eu) which is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-INFRADEV-2019-2, Grant Agreement number: 871115.