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A SEVEN-YEARS BASED CHARACTERISATION OF AEROSOL LIGHT SCATTERING PROPERTIES AT CENTRAL EUROPEAN RURAL SITE: VARIABILITY AND SOURCE APPORTIONMENT

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Keywords: Aerosol optical properties, Scattering and backscattering coefficient, Climate change, Nephelometer, Long-term measurement, Source apportionment

INTRODUCTION

Atmospheric aerosols have a significant impact on the radiative forcing of Earth's climate, either directly through aerosol radiative interactions (ARIs), i.e., scattering or absorption of incoming solar and outgoing infrared radiation, or indirectly through aerosol-cloud interactions (ACIs) (Boucher, 2015; IPCC, 2013; Luoma et al., 2019; Ramanathan et al., 2001). The aerosol radiative forcing of the direct effect consists of a warming effect and a cooling effect (Boucher et al., 2013; Charlson et al., 1992). The predominant cooling effect results from the scattering of radiation by certain species of atmospheric aerosols (including sea salts, nitrates, sulfates, mineral and organic matter, etc.) that reduce the amount of solar radiation reaching the Earth's surface. This phenomenon offsets the greenhouse effect and alters the radiation balance (Pandolfi et al., 2018).

According to a number of studies, radiative forcing by aerosols remains one of the main sources of uncertainty in climate models estimations due to the strong spatial and temporal variations in chemical and physical properties of aerosols, their short lifetime compared to greenhouse gases, and diversity of aerosol sources (Boucher, 2015; Charlson et al., 1992; Lee et al., 2016; Luoma et al., 2019). These studies are important for a better understanding of local and long-range transport of both anthropogenic pollutants and natural aerosols and for unbiased long-term trends.

The aim of this study is to focus on the temporal variations of light-scattering properties of aerosols at a rural background 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), the backscattering ratio (b), and the asymmetry factor (g), are characterized considering different time scales (annual, seasonal, monthly, or diurnal) based on long-term measurement. The optical properties were compared with meteorological conditions (fog, cloudiness); the concentrations of gaseous pollutants such as NO_x and SO₂ were inspected as well as potential sources of atmospheric aerosols. In addition, radiative forcing, and the influence of other meteorological conditions (e.g., height of planetary boundary layer), chemical

composition and particle size distribution at the National Atmospheric Observatory Košetice (NAOK) are being further investigated to better understand the direct effects of aerosols on the local climate.

METHODS

We measured σ_{sp} and σ_{bsp} at three wavelengths (450, 550, and 700 nm) using the Integrating Nephelometer TSI 3563, equipped with a PM10 inlet and a Nafion drier. Measurements were performed 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 Czech Republic. Sampling was conducted at five-minute intervals from August 2012 to February 2013, and then at one-minute intervals till December 2019. All data were processed according to the standard monitoring procedure of the European Monitoring and Evaluation Program (EBAS-EMEP). Periodic control calibrations were performed with CO₂ gas and filtered air to ensure credible results. Moreover, the instrument took part in regular intercomparison exercises organized by ECAC at TROPOS.

Data processing and statistical analysis were performed using R software version 4.1.0. Data were referenced to standard conditions, *SAE* was calculated and used for correction of nonideal light source illumination and cutoff error in the near forward (0 ° -10 °) and near backward (170 ° -180 °) directions using correction factor *C* (Anderson and Ogren, 1998; Massoli et al., 2009; Müller et al., 2009). Later, *b* and *g* were calculated with all temporal variability. Potential source contribution function (PSCF) for estimation of potential aerosol sources was calculated using R software version 4.1.0.

RESULTS

The preliminary results show that the general trend for both σ_{sp} and σ_{bsp} is downward from 2012 to 2019 (Mann-Kendall test; $\alpha=0.05$, $p < 0.05$; except σ_{bsp} at 700 nm). The median slopes of all aerosol light scattering properties are shown in Tab. 1.

SO₂ and NO_x concentrations were correlated with σ_{sp} and σ_{bsp} throughout the period, confirming their contribution to light scattering enhancement (Spearman correlation test; SO₂ and σ_{sp} – $p=1.69e^{-08}$, $\rho=0.557$; NO_x and σ_{sp} – $p=3.91e^{-13}$, $\rho=0.678$; SO₂ and σ_{bsp} – $p=2.29e^{-09}$, $\rho=0.584$; NO_x and σ_{bsp} – $p=5.18e^{-06}$, $\rho=0.464$).

Although σ_{sp} and σ_{bsp} decreased over time, *b* showed a positive slope of the median trend line, indicating a more efficient light backscattering with time. The possible explanation is that the chemical composition of the aerosol changed in favor of cooling (relative decrease in organic/elemental carbon concentration ratio and more scattering chemical species); therefore, aerosol particles become more effective in backscattering at lower aerosol concentrations.

Relative shift from higher to lower *SAE* values (shift to bigger particles) could be an indication of effective policies against industrial/residential emissions. The smallest particles are mainly produced directly at the source (e.g., combustion, heating) or as a product of secondary aerosol (SOA) formation. The trend leading to smaller *SAE* values signals a shift toward larger particles that may be produced by reactions of pre-existing particles in the atmosphere (so-called aerosol aging). Furthermore, long-range transport of aerosol particles might have become a more significant source of aerosol particles in NAOK.

As can be seen in Fig. 1 that σ_{sp} (and σ_{bsp}) reached higher values in the cold season than in the summer. Both σ_{sp} and σ_{bsp} dominated in February, March, and November; 63.2,

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58.7, 58.0 Mm^{-1} and 8.0, 7.2 and 7.0 Mm^{-1} , respectively. This phenomenon is probably related to the higher aerosol load in winter due to higher energy consumption, poorer dispersion of pollutants, and lower altitude of the planetary mixing layer.

Tab. 1: Slopes of the annual median trends found for aerosol light scattering properties throughout 2012-2019 period.

	450 nm	550 nm	700 nm
σ_{sp} [Mm^{-1} /year]	-3.22	-2.50	- 1.03
σ_{bsp} [Mm^{-1} /year]	-0.18	-0.18	-0.09
SAE [/ year]	-0.002	-0.010	-0.018
b [/ year]	0.015	0.012	0.013
g [/ year]	-0.027	-0.021	-0.022

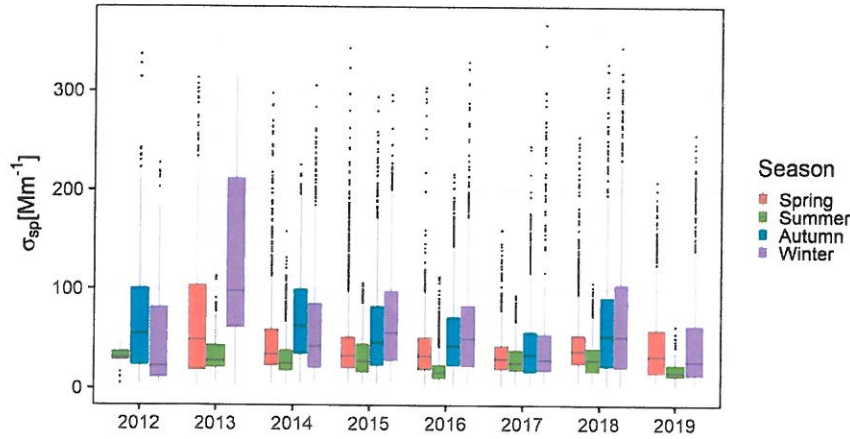


Fig. 1: The annual and seasonal variation of σ_{sp} at 550 nm. Black line represents median, solid black circles represent outliers.

The elevated levels of SAE in summer might indicate enhanced formation of SOA during the photochemically active season. SAE levels reached the highest monthly value 2.09 in July. This is supported by the correlation of the increase in isoprene concentration at NAOK in summer as well as by the results published in Mbengue et al. (2021, 2020). In contrast, particles remain longer in the atmosphere during the winter season and can overcome several chemical reactions during aerosol aging and thus grow, reaching the lowest monthly value of SAE in February; 1.62.

The overall annual trend of b is upward from 2012 to 2019 (Tab. 1) what could be explained by possible partial change in particle size and chemical composition of aerosols toward more backscattering species, e.g., secondary organic aerosols or transport of oxidized/sea-salt aerosol particles during summer, which is mainly pronounced in summer period. The maximum was observed in May, June, and July; 0.16 in all cases, and the minimum in November; 0.12.

As the g ranges from -1 to 1 (total backscattering to total forward scattering), the overall lowering of the g value shows a shift to stronger backscattering. Maximum values of g were observed in November, February, and March; 0.6 in all cases. Since higher values

of g represent a dominance of forward scattering, this suggests less backward scattering species in winter, e.g., aerosol mixtures containing carbonaceous substances. However, median values of g remain in the positive values for the whole inspected period indicating prevalence of forward scattering of light, which is in agreement with theoretical predictions.

The diurnal variations of σ_{sp} and σ_{bsp} were downward from midnight and reached their minimum around noon (29.6 Mm^{-1} and 4.3 Mm^{-1} , respectively) and started to increase to reach their maximum at 9 p.m. (39.58 Mm^{-1} and 5.6 Mm^{-1} , respectively). The main reason for this is the higher stability of the atmosphere and the lower planetary boundary layer during nighttime.

The daily variation of SAE included two maximum peaks (the smallest size), one in the early morning at 3-5 a.m.; 1.86 and the second at 1 p.m.; 1.85. The lowest SAE values were observed daily between 6 am and 8 a.m., indicating the occurrence of largest particles during this part of the day. No visible variations were observed for b during the week, and the median remained stable at 0.14.

The PSCF visualization of aerosol potential source identification is shown in Fig. 2. PSCF showed a higher probability of source location identification of the observed values

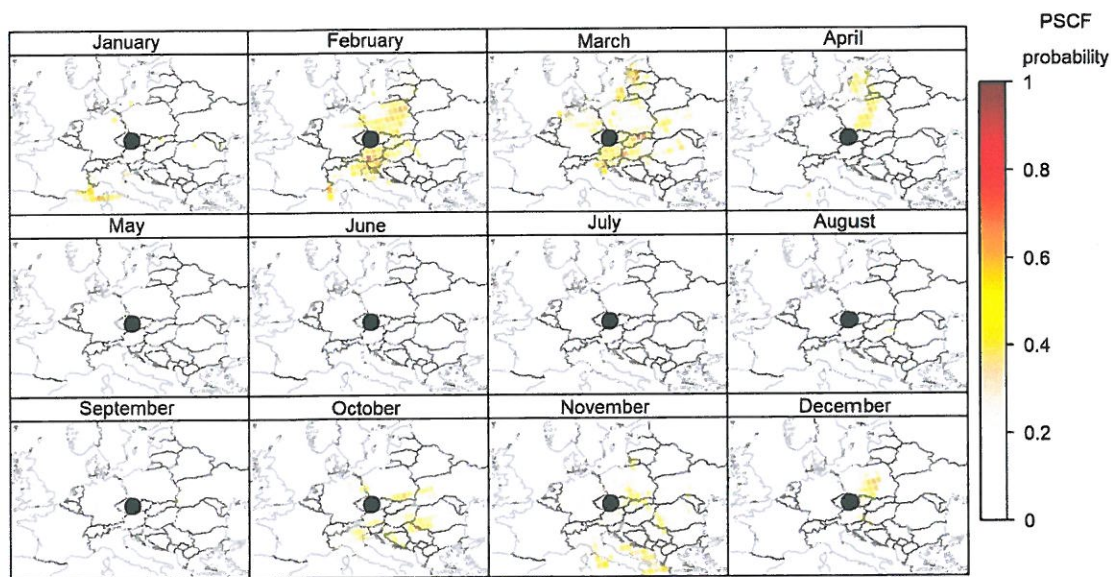


Fig. 2: PSCF of σ_{sp} at 550 nm calculated separately for different months with monthly 75th percentile as the limit value. The location of NAOK is marked with black circle.

over 75th percentile of σ_{sp} during late autumn, winter, and early spring. This probability correlates with higher σ_{sp} and σ_{bsp} levels during cold season. As the probability is not dense in any region, the results may be more by synoptic situation than potential sources of aerosols.

To compare AOP with cloudiness, we defined four categories according to WMO definition – fine F (no clouds), partly cloudy PC, cloudy C and overcast O days (completely covered sky without breaks).

Higher values of σ_{sp} and σ_{bsp} observed during O days could be caused by compressed boundary layer. However, we expect more precipitation during O days, thus removing aerosol particles from the atmosphere, which was not proven in this study. For F days, the hypothesis suggests higher solar activity and photooxidative processes in the

atmosphere causing higher production of secondary organic aerosols. At 550 nm, the σ_{sp} medians were 36.46 Mm⁻¹, 28.81 Mm⁻¹, 28.12 Mm⁻¹ and 41.42 Mm⁻¹ and the σ_{bsp} medians 5.47 Mm⁻¹, 4.61 Mm⁻¹, 4.41 Mm⁻¹ and 5.52 Mm⁻¹ for days F, PC, C, and O, respectively.

A study of fog influence on AOP showed increased scattering potential of aerosols during days without the fog, because the multiple-scattering of light on particles might occur during the foggy days causing decreased intensity of measured light coming to the instrumental cell.

CONCLUSIONS

In this study, we focused on temporal variations of long-term measurement of AOP at a rural site in Central Europe, as well as on the comparison with different meteorological conditions, concentration of gaseous pollutants, and source apportionment.

The overall trend of σ_{sp} and σ_{bsp} was decreasing from 2012 to 2019. Similar decreasing trend was observed for *SAE*, indicating shift to relatively bigger particles, increased *b* and decreased *g* suggests increased efficiency of backscattering at lower concentration, bigger particle size and possible change in chemical composition. Elevated *SAE* in the summer implies SOA formation, decreased *SAE* in winter denotes aerosol aging, and higher atmospheric stability. Decreased *b* in winter suggests the occurrence of atmospheric particles with lower backscattering potential, e.g., carbonaceous aerosols originating from different types of combustion. The backscattering potential was decreased during fog events (multiple scattering of light). The source identification did not show a high probability of occurrence in any of the regions, suggesting the prevalent influence of the synoptic situation over Europe more than the potential sources of aerosols.

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