

# 1 Hygroscopic enhancement of suburban aerosol light scattering measured using a 2 single-nephelometer system in Central Europe

3 Lenka Suchánková<sup>1,2,3</sup>, Jakub Ondráček<sup>1</sup>, Naděžda Zíková<sup>1</sup>, Petr Roztočil<sup>1</sup>, Petr Vodička<sup>1</sup>, Roman  
4 Prokeš<sup>2,3</sup>, Ivan Holoubek<sup>2,3+</sup>, Vladimír Ždímal<sup>1</sup>

5 <sup>1</sup>Institute of Chemical Process Fundamentals of the Czech Academy of Sciences, Prague, 165 00, Czech Republic

6 <sup>2</sup>Global Change Research Institute of the Czech Academy of Sciences, Brno, 603 00, Czech Republic

7 <sup>3</sup>RECETOX, Faculty of Science, Masaryk University, Brno, 611 37, Czech Republic

8 <sup>+</sup> deceased

9 *Correspondence to:* Lenka Suchánková ([suchankova@icpf.cas.cz](mailto:suchankova@icpf.cas.cz))

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12 **Abstract:** Most atmospheric aerosol particles are hygroscopic, meaning they absorb water from the surrounding air, altering  
13 their size, shape, overall chemistry, refractive index, and thus light-scattering properties — an effect with important  
14 implications for Earth's radiative balance. The scattering enhancement factor,  $f(\text{RH})$ , and backscattering enhancement factor,  
15  $f(\text{RH})_{\text{bsp}}$ , quantify the increase in light scattering under elevated relative humidity (RH). These parameters are typically  
16 measured using two nephelometers operating under dry ( $\text{RH} < 40\%$ ) and humidified ( $\text{RH} > 80\%$ ) conditions, a method prone  
17 to inter-instrument uncertainties. This study presents a single-nephelometer system that reduces measurement uncertainty  
18 associated with inter-instrument comparison and enables the study of aerosol hygroscopic behavior in the inadequately  
19 represented European urban environment. The system was deployed at a suburban site in Prague, Suchdol, Czech Republic,  
20 from November 2022 to August 2023. Results revealed low aerosol hygroscopicity, likely due to a well-mixed aerosol  
21 population dominated by black and brown carbon. Both enhancement factors peaked in spring, coinciding with the enhanced  
22 formation of secondary aerosols and particle growth, which modified aerosol size distributions and hygroscopicity. Low  
23 hygroscopic enhancement values in summer reflected a composition shift toward black carbon-dominated aerosols from traffic  
24 emissions, with particle growth being disrupted, potentially due to the structural compaction of black carbon aggregates under  
25 high RH. While  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  generally increased with decreasing concentrations of light-absorbing particles, organic  
26 carbon, particularly its most volatile fractions, significantly enhanced aerosol hygroscopicity in the urban environment. Despite  
27 overall low aerosol hygroscopicity, increased RH significantly influenced aerosol climate-relevant optical properties

## 28 1. Introduction

29 Atmospheric aerosols play a critical role in the Earth's energy budget through direct aerosol-radiation interactions (ARI)  
30 by the scattering and absorption of short- and long-wave radiation and indirect aerosol-cloud interactions (ACI) by changes in  
31 the microphysical and radiative properties of clouds, respectively (Boucher, 2015; IPCC, 2021). The Sixth Assessment Report  
32 of the IPCC estimated the total aerosol effective radiative forcing (ERF) to be  $-1.1$  [ $-1.7$  to  $-0.4$ ]  $\text{W m}^{-2}$  over 1750-2019 (Foster  
33 et al., 2023). Despite growing research on the aerosol radiative effects (e.g., Toll et al., 2019; Williams et al., 2022; Zhang et  
34 al., 2025 and references herein), aerosol ERF remains the most significant uncertainty in climate models due to the high spatial  
35 and temporal variability of aerosol properties, limited understanding of pre-industrial aerosol conditions, and the indirect  
36 aerosol-induced changes in the atmosphere (Carslaw et al., 2017; Kahn et al., 2023; Watson-Parris and Smith, 2022).

37 Hygroscopicity, defined as the ability of aerosol particles to attract and absorb moisture from the surrounding  
38 environment, critically alters particle size, shape, and refractive index (Burgos et al., 2019; Titos et al., 2021) and impacts the  
39 angular distribution of scattered light and thus aerosol optical properties (Fierz-Schmidhauser et al., 2010; Zieger et al., 2015).

40 Since the globally measured long-term in situ aerosol measurements are standardized below 40 % relative humidity (RH)  
41 (WMO/GAW, 2016), these "dry" conditions do not reflect the real atmosphere, leading to an inadequate understanding of  
42 aerosol water uptake, which contributes to significant uncertainties affecting aerosol climate effects (Burgos et al., 2020; Myhre  
43 et al., 2013; Ray et al., 2024).

44 The light scattering enhancement due to humidity can be expressed by the light scattering enhancement factor  $f(\text{RH})$   
45 as in Eq. (1):

$$46 \quad f(\text{RH}) = \frac{\sigma_{\text{sp}}(\text{RH}, \lambda)}{\sigma_{\text{sp}}(\text{RH}_{\text{dry}}, \lambda)}, \quad (1)$$

47 where  $\sigma_{\text{sp}}(\text{RH}, \lambda)$  and  $\sigma_{\text{sp}}(\text{RH}_{\text{dry}}, \lambda)$  denote total scattering coefficients under elevated RH conditions and dry conditions  
48 measured at the same wavelength  $\lambda$ , respectively (Covert et al., 1972). A similar formulation applies for backscattering,  
49  $f(\text{RH})_{\text{bsp}}$  (Titos et al., 2021).

50 Several approaches to investigate  $f(\text{RH})$  have been proposed. Tandem-humidified nephelometer systems occurred in the  
51 1960s and have undergone substantial innovations since then (Pilat and Charlson, 1966). These systems consist of one  
52 nephelometer measuring under dry conditions and a second nephelometer measuring a humidified aerosol sample. Two main  
53 instrumental set-ups were identified in the 26 tandem-humidified nephelometer measurements from ground-based sites  
54 worldwide (Burgos et al., 2019). The "NOAA design" directs aerosol through a first dry and later humidified nephelometer  
55 (e.g., Doherty, 2005; Liu and Li, 2018), while the "PSI design" splits the aerosol into parallel dry and humidified paths (e.g.,  
56 Zieger et al., 2015, 2014). Both systems used an RH scanning regime (20 to 95 % RH) for the humidified nephelometer (Titos  
57 et al., 2016).

58 Müller et al. (2011) showed that after correction for angular truncation errors, the total scattering coefficients  $\sigma_{\text{sp}}$  and  
59 backscattering coefficients  $\sigma_{\text{bsp}}$  at 450, 525 and 635 nm measured by the Ecotech Aurora 3000 against the referenced integrating  
60 nephelometer TSI 3563 in laboratory conditions differ by 2-5 % and 1-11 %, respectively. The discrepancies observed in total  
61 scattering were consistent with the calibration uncertainties. The experimental set-ups comprising two or more instruments  
62 could introduce additional uncertainty to the resulting data, considering different sampling lines for nephelometers, non-  
63 symmetrical apparatus, or the critical measurement part under highly humid conditions (Anderson et al., 1996).

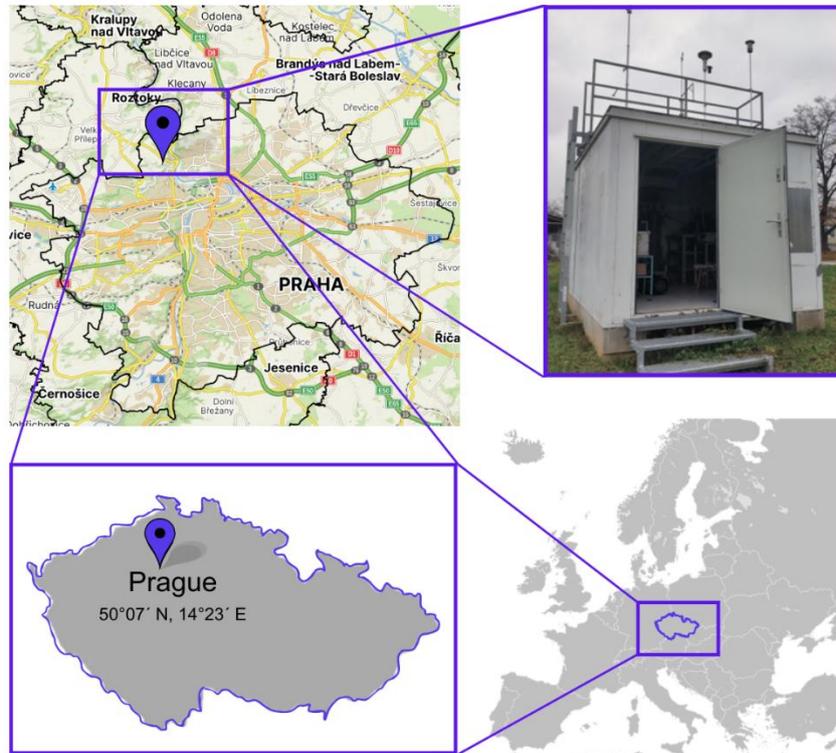
64 Thus, this study presents a single-nephelometer system to partially reduce uncertainties in the  $f(\text{RH})$  estimation arising  
65 from the comparison of two instruments and to investigate ambient aerosol particles' light scattering hygroscopic behavior at  
66 the suburban site. Orozco et al. (2016) examined aerosol hygroscopicity using a dryer-humidifier system coupled to a TSI  
67 3563 nephelometer in urban/suburban environments in North America. However, to the best of our knowledge, only one study  
68 has specifically investigated aerosol light-scattering enhancement in a European urban/suburban environment (Titos et al.,  
69 2014). Therefore, this study provides a unique insight into  $f(\text{RH})$  and light-scattering enhancement of aerosols in a European  
70 suburban context using a single-nephelometer approach. Moreover, the presented approach enabled the reliable measurement  
71 of hygroscopic changes in the aerosol light backscattering  $f(\text{RH})_{\text{bsp}}$ , inadequately represented in the literature.

## 72 2. Materials and Methods

### 73 2.1. Description of the site

74 The instrumentation set-up was developed and tested at the Institute of Chemical Process Fundamentals (ICPF) of the  
75 Czech Academy of Sciences in Prague, Czech Republic. The ICPF also runs a Suchdol atmospheric station located on the  
76 institute campus (50° 7' 35" N, 14° 23' 5" E, 277 m a.s.l., Fig. 1). The station is a suburban site and an Aerosol In Situ National  
77 Facility (AIS NF) of the ACTRIS ERIC (Aerosols, Clouds, and Trace gases Research InfraStructure, European Research  
78 Infrastructure Consortium; <https://www.actris.net/>). The aerosol instruments are positioned within the sampling container, with  
79 the sampling heads situated approximately 4 meters above the ground.

80 The station is located at the periphery of the plateau above the capital, Prague (1.37 million citizens in 2025), 5 km from  
81 the city center. The site is surrounded by residential housing, utilizing gas as the primary energy source for heating (80 %),  
82 while the remainder utilizes electricity (16 %), community heating (2 %), or burns solid and liquid fuels (less than 2 %) (Český  
83 statistický úřad, 2021). The nearest road is situated at a distance of 250 m (10,000-15,000 cars per day, Vodička et al., 2013),  
84 but no major road is located within 1 km of the site. The Václav Havel airport is situated 9 km SW of the site. The agricultural  
85 fields are located within a 2 km radius to the west. The predominant wind direction at the site is from the WSW (mainly  
86 summer and winter), with a notable influence of SE during winter and NW during spring (Fig. S.1). The measurement  
87 campaign was conducted from 15 November 2022, to 19 August 2023.

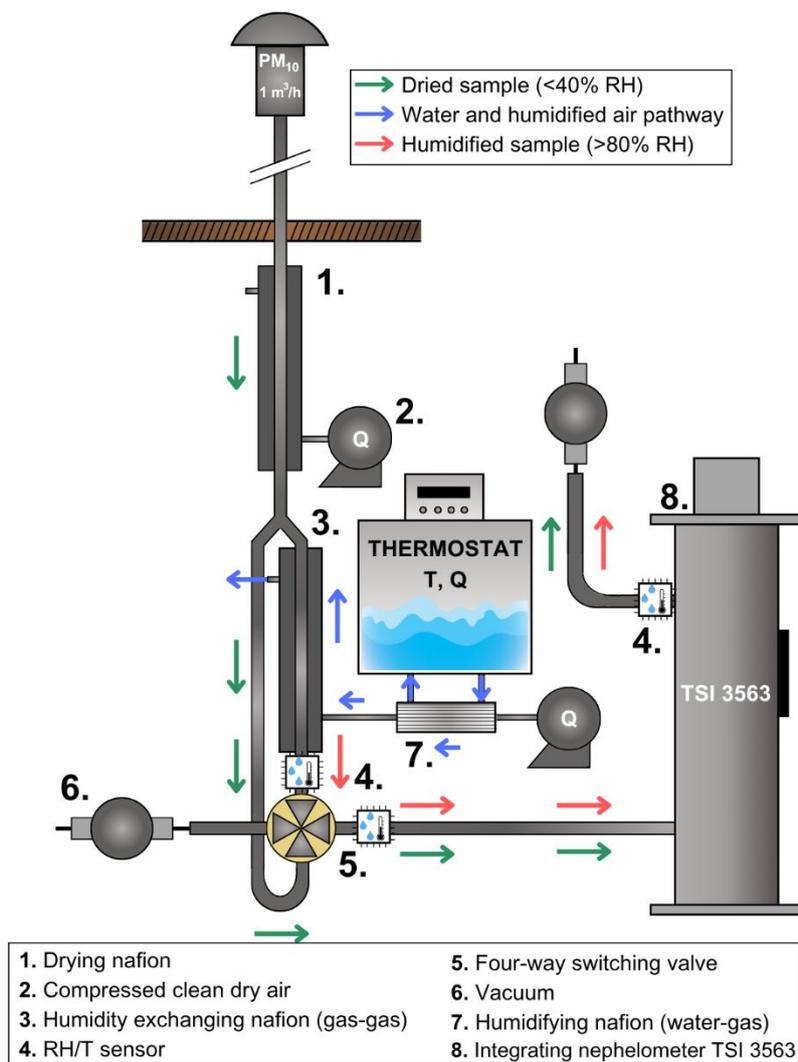


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89 **Figure 1: Location of Suchdol ACTRIS ERIC AIS NF site within Prague, Czech Republic. Map source: © Seznam.cz, a.s.**

## 90 2.2. The single-nephelometer instrumentation

91 Aerosol particles were sampled through a PM<sub>10</sub> sampling head (Leckel, GmbH) and subsequently dried by a custom-built  
92 Nafion dryer (MD-700-24, Permapure) to achieve the RH level below 40 % (Fig. 2, No. 1). The total aerosol flow of 10 lpm  
93 was divided equally between two parallel sampling lines: a dry sampling line (5 lpm) and a humidified sampling line (5 lpm).  
94 The dry sample was passed through the sampling system to the integrating nephelometer (TSI 3563) without additional  
95 adjustments. The other line led to the second Nafion membrane (MD-700-24, Permapure), functioning as a water exchange  
96 medium, facilitating counter-current mass transfer between humidified particle-free air and the aerosol sample (Fig. 2, No. 3).  
97 This humidification process aimed to achieve a sample RH  $\geq$  80 %.



98

99 **Figure 2: A design of the single-nephelometer set-up system for studying aerosol hygroscopic behavior.**

100 Humid particle-free air was produced in the closed circulation system (Fig. 2, blue arrows). Demineralized water,  
 101 heated in a controlled manner by the thermostat (up to 33 °C), was directed to the bundle Nafion membrane (FC100,  
 102 Permapure) (Fig. 2, No. 7), where mass transfer between water (in channels) and the dry particle-free air (outside the channels)  
 103 occurred. The excessive water was later returned to the thermostat, and the humid particle-free air flowed into the humidity-  
 104 exchanging Nafion membrane (Fig. 2, No. 3). The temperature of the demineralized water and the flow rate of the humid  
 105 particle-free air were regulated to achieve the desired RH level in the humidity-exchanging Nafion. An RH/T sensor (HYT939,  
 106 Innovative Sensor Technology, AG) was installed before the switching valve to control the RH in the humidified sample. Since  
 107 the RH sensor inside the measurement cell was not sufficiently accurate, additional RH/T sensors were positioned immediately  
 108 in front of the inlet and after the outlet of the instrument (Fig. 2, No. 4) to control the RH dynamics and to calculate the dew  
 109 point temperature  $T_{dew}$ , which was used to estimate the real RH of the sample (see Chapter 2.4 Data treatment). All RH/T  
 110 sensors were calibrated against a standard thermometer (F250 MKII, Automatic System Laboratories) and a dew point mirror  
 111 (CMH2, Alpha Moisture Systems) at the beginning of the campaign.

112 Every 60 minutes, the four-way switching valve (Fig. 2, No. 5) automatically directed either the dry or the humidified  
 113 sample to the TSI 3563 integrating nephelometer (Fig. 2, No. 8). First, a 10-minute conditioning period was initiated to reach  
 114 the target RH of the sample, followed by a 50-minute measurement period as determined and optimized by pilot testing. During  
 115 the dry sample measurement, the automatic switching valve allowed the dry sample to flow directly to the nephelometer, and

116 the humidified sample was directed to the exhaust (Fig. 2, No. 6). And vice versa, when the humidified aerosol sample was  
117 sampled to the nephelometer, the dry sample was discarded.

118 The thermostat temperature and the flow rate of the humidified particle-free air were checked regularly to ensure  
119 proper humidification of the sample and to prevent condensation inside the instrument. All parts sensitive to changes in RH  
120 (humidity-exchanging Nafion, tubing within the closed humidity circuit, and inlet tubing to the nephelometer) were insulated  
121 to prevent heat losses and water condensation.

122 Upon reaching the nephelometer measuring cell, a dry or humidified aerosol sample was illuminated with a halogen  
123 lamp at an angle range of 7°–170°. The scattered light passed through three band-pass filters and was detected in  
124 photomultiplier tubes (PMT) at 450, 550, and 700 nm wavelengths. The resulting total scattering and backscattering  
125 coefficients ( $\sigma_{sp}$  and  $\sigma_{bsp}$ ) with a time resolution of 1 minute.

126 The nephelometer was calibrated twice a day with particle-free air and fully calibrated every 2–3 months with CO<sub>2</sub> as the high-  
127 span gas and particle-free air as the low-span gas, always in the dry measurement regime. The continuous dry measurement  
128 was performed approximately once a month (overnight) to avoid water condensation inside the instrument.

129 Particle losses in the Nafion membranes and the 4-way valve are not expected to significantly influence the reported  
130 scattering measurements, as diffusional losses affect optically negligible ultrafine particles and inertial losses primarily affect  
131 coarse particles that are scarce in urban air. Urban aerosol scattering is dominated by accumulation-mode particles (< 1  $\mu\text{m}$ )  
132 (Held et al., 2008; Wu and Boor, 2021), for which calculated valve losses are minimal.

133 While this approach reduces uncertainties in the  $f(\text{RH})$  estimation arising from the comparison of two instruments, it  
134 should be noted that such a measurement approach relies on the reduced time resolution compared to the dual nephelometer  
135 setups (1-hour intervals), which may introduce additional uncertainty on short timescales, e.g., the influence of episodic  
136 extreme pollution events. This limitation was partly addressed in the Chapter Data treatment. Moreover, another limitation of  
137 this setup originates from a lack of parallel measurement of dry and wet aerosol properties, which rules out the hygroscopic  
138 scanning, humidogram analyses, and the precise recalculation of  $f(\text{RH})$  at the given RH. The statistical overview of RH and  
139 temperature during the measurement campaign for both humidified and dry mode are shown in Table S.2 in Supplementary  
140 Materials.

### 141 2.3. Auxiliary measurements

142 The Mobility Particle Size Spectrometer (MPSS) measured the aerosol particle number concentration with a time  
143 resolution of 5 minutes, using a custom-built Differential Mobility Analyzer (DMA, both TROPOS, Germany), positive high-  
144 voltage power supply, and Condensational Particle Counter (CPC 3772, TSI). The MPSS ranged from 10 to 800 nm, 32 size  
145 channels per decade, with data further subdivided into size modes: 8–100 nm, 100–200 nm, 200–500 nm, and particles above  
146 500 nm for the analysis. An additional total count CPC (3750, TSI) measured the total particle number concentration of  
147 particles larger with  $d_p(50)$  at 10 nm.

148 Elemental and organic carbon (EC and OC) concentrations were measured from November 2022 to July 2023 using  
149 a semi-online field analyzer from Sunset Laboratory Inc. (USA) (Bauer et al., 2009). The analyzer was connected to a PM<sub>1</sub>  
150 (November – December 2022) and PM<sub>2.5</sub> (rest of the period) inlet with a flow rate of 8 lpm. Samples were collected at 2-hour  
151 intervals on a quartz fiber filter and analyzed according to the shortened EUSAAR2 protocol (Cavalli et al., 2010). Each  
152 measurement was corrected for charring, and the RTCalc726 software automatically determined the split point between EC  
153 and OC using a linear fit of laser and temperature corrections. OC was divided into fractions based on temperature. The most  
154 volatile fractions, OC1 and OC2, volatilize at 200 °C and 300 °C and are commonly present in fresh vehicle exhaust, biomass  
155 burning, and coal combustion (Shen et al., 2025; Vodička et al., 2015). OC3 and OC4 subfractions volatilize at 450 °C and 650  
156 °C and represent less volatile fractions of OC with higher molecular weights and are associated with chemical aging and the  
157 products of photochemical reactions (Aswini et al., 2019; Shen et al., 2025). The instrument was equipped with a parallel

158 carbon plate denuder to eliminate volatile organic compounds and prevent positive bias in OC measurements. Instrument  
159 blanks were recorded daily at midnight.

160 A dual-spot multiwavelength aethalometer (Model AE33, Magee Scientific, USA, 2018) continuously measured light  
161 attenuation by particles at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm). The dual-spot technology enables  
162 real-time compensation for filter loading. Particles were sampled through the PM<sub>10</sub> sampling head (Leckel GmbH) at a flow  
163 rate of 5 lpm, dried in a custom-made Nafion dryer (TROPOS, Leipzig, Germany), and deposited onto tetrafluoroethylene  
164 (TFE) coated glass filter tape. Light transmission through the deposited sample is measured and compared to the blank filter  
165 tape spot as a reference, converting the optical absorbance into an equivalent black carbon concentration (eBC,  $\mu\text{g m}^{-3}$ ) data.  
166 The data was automatically corrected by the multi-scattering correction factor C (1.39 for the recommended filter tape M8060).  
167 Furthermore, the wavelength-dependent mass absorption cross-section (MAC) factors were used for the eBC conversion to  
168 the absorption coefficients  $\sigma_{\text{ap}}$  (Drinovec et al., 2015; Müller and Fiebig, 2021; Savadkoohi et al., 2025). The wavelength-  
169 dependent MAC values were adopted from the AE33 manual (e.g.,  $\text{MAC} = 7.77 \text{ m}^2 \text{ g}^{-1}$  for 880 nm) (Magee Scientific, 2018).  
170 The  $\sigma_{\text{ap}}$  values were additionally standardized to STP conditions (273.15 K, 1013.25 hPa) and divided by the harmonization  
171 factor H\* (1.76 for the recommended filter tape M8060), which compensates for the differences between the predefined multi-  
172 scattering correction factor C and corrections in the Aethalometer firmware set by the manufacturer (Müller and Fiebig, 2021;  
173 Savadkoohi et al., 2024, 2025).

174 Ambient temperature (T), RH, wind speed (WS), wind direction (WD), global solar radiation (GLRD), and ozone (O<sub>3</sub>)  
175 concentration were measured hourly in the Czech Hydrometeorological Institute container located next to the ACTRIS AIS  
176 container with all the aerosol instruments.

## 177 2.4. Data treatment

### 178 2.4.1. Humidified nephelometer system

179 Four subsystems working simultaneously were needed to obtain valid datasets from the measurements: nephelometer  
180 measurement, automatic switching between dry and humid measurements, humidification of the sample, and RH/T sensors.

181 The TSI 3563 integrating nephelometer data set was processed according to the EMEP Standard Operating Procedure.  
182 The raw  $\sigma_{\text{sp}}$  and  $\sigma_{\text{bsp}}$  data at all wavelengths were validated (removal of invalid, missing, and calibration data). Values below  
183 the limit of detection LOD ( $0.3 \text{ Mm}^{-1}$  for TSI 3563) were replaced by LOD/2 values, corrected for a truncation error according  
184 to Anderson and Ogren (1998), and standardized to STP conditions (273.15 K, 1013.25 hPa).

185 The automatic four-way switching valve was controlled using a custom-made LabVIEW program. The resulting data  
186 set was paired with the nephelometer data set to identify dry and humidified measurements.

187 All datasets produced by the RH/T sensors regulating the thermostat temperature and the flow of humidified particle-free air  
188 to achieve the desired RH of the sample were also recorded using a custom-made LabVIEW program.

189 Temperature and RH data from all three sensors were corrected using calibration curves derived from comparing with  
190 the referenced thermometer and the dew point mirror. The real RH of the sample was derived by assuming that the dew point  
191 temperatures in front of and behind the cell are similar (Ren et al., 2021). The approximated Magnus-Tetens formula (Alduchov  
192 and Eskridge, 1997) was used to calculate the dew point temperatures of both RH/T and from the mean  $T_{\text{dew}}$ , the saturation  
193 vapor pressure,  $p_{\text{sat}}$ , at  $T_{\text{dew}}$  was calculated as in Eq. (2):

$$194 \quad p_{\text{sat}} = a * e^{\frac{b * T_{\text{dew}}}{T_{\text{dew}} + c}}, \quad (2)$$

195 where a, b, and c are empirical constants derived from the experimental data: a = 6.112 hPa, b = 17.67, and c = 243.51 (b and  
196 c are dimensionless);  $T_{\text{dew}}$  in °C.

197 A similar approach was used to calculate the saturation vapor pressure at the temperature in the measuring cell,  $T_{\text{cell}} - p_{\text{cell}}$ .

198

The final RH of the sample in the measuring cell,  $RH_{cell}$ , was calculated as follows in Eq. (3):

199

$$RH_{cell} = \left( \frac{p_{sat}}{p_{cell}} \right) * 100. \quad (3)$$

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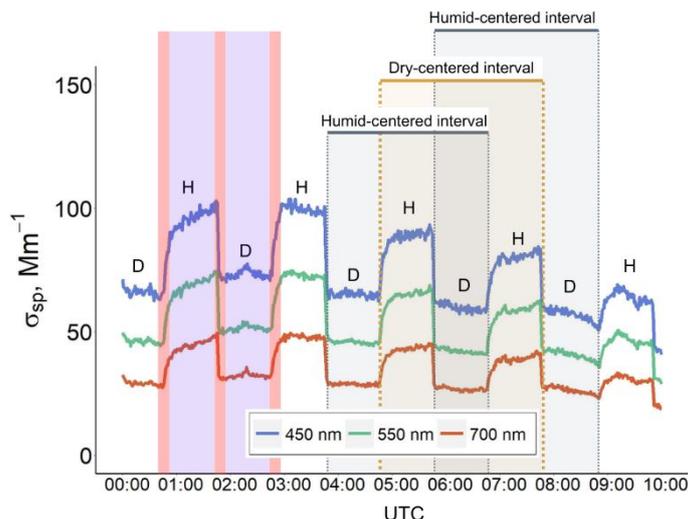
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The  $RH_{cell}$  dataset was later combined with the nephelometer and switching valve data. The  $RH_{cell}$  data, along with the valve position dataset, were used to separate the data into dry ( $RH \leq 40\%$ ) and humidified ( $RH \geq 80\%$ ) datasets. They were also used to separate the conditioning periods, which occurred when switching between dry and humidified modes to reach RH equilibrium in the measurement cell from the actual measurement periods (Fig. 3). Data corresponding to the range of  $40\% < RH < 80\%$ , including the conditioning periods, was discarded (approx. 15% of the raw data). The dry and humidified datasets of aerosol light scattering properties were averaged hourly. The enhancement factors  $f(RH)$  and  $f(RH)_{bsp}$  were calculated to obtain information about light scattering enhancement due to hygroscopicity. In this study,  $f(RH)$  was calculated based on "humid-centered" and "dry-centered" intervals to avoid the influence of possible extreme pollution events at the site (Fig. 3). The humid-centered interval was calculated by dividing the average humidified  $\sigma_{sp}$  value by the mean of two lateral 1-hour averaged dry  $\sigma_{sp}$  (Fig. 3, gray). The dry-centered interval was calculated by dividing the average of two lateral 1-hour averaged humidified  $\sigma_{sp}$  by the 1-hour averaged dry  $\sigma_{sp}$  value (Fig. 3, ochre). Extreme and invalid values were inspected and discarded if necessary. The data coverage of the entire measurement campaign, including individual seasons, as well as the variability of RH and temperature under humid and dry conditions, can be found in Table S.1, Table S.2, and Fig. S.1 in Supplementary Materials.



214

**Figure 3: The example of  $f(RH)$  calculation from a single-nephelometer measurement on December 12, 2022. The D and H symbols indicate "dry" and "humid" measuring intervals of  $\sigma_{sp}$ . The orange intervals on the left represent the preconditioning periods, while the purple intervals represent the actual measurements.**

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#### 2.4.2. Relationship between aerosol scattering and absorption

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219

To describe the spectral dependence of light scattering, the Scattering Ångström Exponent SAE was calculated as (Clarke and Kapustin, 2010):

220

$$SAE_{\lambda_1-\lambda_2} = - \frac{\log \left( \frac{\sigma_{sp}(\lambda_1)}{\sigma_{sp}(\lambda_2)} \right)}{\log \left( \frac{\lambda_1}{\lambda_2} \right)}, \quad (4)$$

221

222

223

where  $\lambda_1$  and  $\lambda_2$  are the wavelengths of light at which  $\sigma_{sp}$  was measured. SAE contains information about aerosol size: SAE values  $< 1$  indicate the predominance of particles in the coarse mode, while SAE values  $\geq 2$  indicate a predominance of the aerosol fine mode (Seinfeld and Pandis, 2006).

224

In addition to SAE,  $\Delta SAE$  was defined (Perrone et al., 2018) in Eq. (5):

225

$$\Delta SAE = SAE_{450-550} - SAE_{550-700}, \quad (5)$$

226 where  $SAE_{450-550}$  is calculated from 450 and 550 nm and  $SAE_{550-700}$  from 550 and 700 nm wavelength pair, respectively.  
 227  $\Delta SAE$  provides insight into the relative contribution of fine and coarse mode particles and whether the particle size  
 228 distribution is mono-, bi-, or multimodal. Positive  $\Delta SAE$  values indicate the presence of two distinct modes—a fine mode  
 229 and a coarser one—while negative  $\Delta SAE$  values suggest the dominance of a single fine particle mode (Perrone et al., 2018).

230 The Absorption Ångström Exponent (AAE), which describes a spectral dependence of light absorption, was  
 231 calculated analogously from  $\sigma_{ap}$  (Mbengue et al., 2021). AAE can provide information on chemical composition: AAE values  
 232  $< 1$  could indicate BC core or non-absorbing coating particles, AAE values around 1 are classified as BC aerosol, while AAE  
 233 values around 2 and higher indicate light absorption in ultraviolet and blue spectral regions, suggesting the presence of organic  
 234 carbon — brown carbon BrC in this study — or mineral dust (Cappa et al., 2016).

235 To estimate the dominant aerosol type at the site, we used the AAE vs. SAE plot from Cappa et al. (2016), which can  
 236 estimate the potential aerosol type without direct information on the chemical composition. For the aerosol type assessment in  
 237 this study, the AAE was calculated at 520–660 nm and the SAE at 450–550 nm. The AAE vs. SAE plots were additionally  
 238 color-coded with the Single Scattering Albedo (SSA), the ratio of the aerosol light scattering, and the total aerosol light  
 239 extinction (light scattering plus absorption) at the predefined wavelength  $\lambda$  defined in Eq. (6):

$$240 \quad SSA_{\lambda} = \frac{\sigma_{sp}(\lambda)}{\sigma_{sp}(\lambda) + \sigma_{ap}(\lambda)}. \quad (6)$$

241 Sites predominantly influenced by aerosol scattering (clean marine or remote Arctic sites) exhibit SSA values close to 1, while  
 242 anthropogenically influenced sites exhibit significantly lower SSA (Pandolfi et al., 2018). In this study, also the humidified  
 243 equivalent of SSA was calculated from dry measurements of  $\sigma_{ap}$  (RH  $< 40$  %) and humidified  $\sigma_{sp}$  (RH  $> 80$  %).

244 The asymmetry factor  $g$  describes the angular distribution of the scattered light and is defined as the average cosine  
 245 of the angle between the incident light and the scattered beam  $\theta$ , weighted by the probability of scattering for each possible  
 246 angle. Based on the Henyey-Greenstein approximation (Andrews et al., 2006; Wiscombe and Grams, 1976):

$$247 \quad g_{\lambda} = -7.143889b_{\lambda}^3 + 7.464439b_{\lambda}^2 - 3.96356b_{\lambda} + 0.9893, \quad (7)$$

248 where  $b$  is the hemispheric backscattering ratio.  $g$  ranged from -1 for completely back-scattered light to 1 for completely  
 249 forward-scattered light and is one of the essential inputs for the radiative transfer models.

250 The hemispheric backscattering ratio  $b$  denotes the fraction of light scattered back to the upper hemisphere of the  
 251 particle and the total scattered light and can be measured directly from the optical instrument without knowledge of the  
 252 scattering phase function (ranging from 0 to 1) calculated following Eq. (8):

$$253 \quad b_{\lambda} = \frac{\sigma_{bsp}(\lambda)}{\sigma_{sp}(\lambda)} \quad (8)$$

254  $g$  and  $b$  are particularly useful for distinguishing aerosol types and assessing their radiative impacts, as backscattering plays a  
 255 critical role in determining the cooling efficiency of atmospheric particles.

### 256 **2.4.3. Back trajectory analysis**

257 The dry optical properties data were paired with a cluster analysis of back trajectories calculated using the  
 258 HYSPLIT\_4 model from the NOAA Air Resources Laboratory to understand the sources of distinct aerosol types better. The  
 259 global data assimilation system (GDAS) at  $1^{\circ} \times 1^{\circ}$  resolution (Draxler and Hess, 1998; Stein et al., 2015) was used as  
 260 meteorology input, and 72-hour air mass back trajectories arriving at 200 m a.g.l. were calculated every 6 hours. The number  
 261 of clusters was estimated based on total spatial variance.

262 **3. Results and discussion**

263 **3.1. Light scattering properties**

264 The overall median value of dry  $\sigma_{sp}$  at 550 nm (text refers to the overall measurement at  $\lambda = 550$  nm unless stated otherwise)  
 265 was 28.45  $Mm^{-1}$  at the studied site, corresponding to the range of values observed at urban and suburban sites: 14.83  $Mm^{-1}$  at  
 266 SIRTa (FR), 47.39  $Mm^{-1}$  in Athens (GR), 39.83  $Mm^{-1}$  in Lecce (IT), 18.04  $Mm^{-1}$  and 43.14  $Mm^{-1}$  in Madrid and Granada (ES)  
 267 (Donateo et al., 2020; Pandolfi et al., 2018).

268 The dry  $SAE_{450-700}$  value of 1.65, together with a positive median dry  $\Delta SAE$  (Table 1), indicated a predominantly  
 269 fine-mode aerosol population, with spectral curvature suggesting the presence of a secondary mode associated with larger  
 270 particles, likely from aging or mixing processes, similarly to Athens and Granada ( $SAE_{450-700}$  of 1.6 and 1.69), while in Lecce,  
 271 the fine particle mode dominated ( $SAE_{450-700}$  of 1.84) (Donateo et al., 2020; Pandolfi et al., 2018). The dry  $b$  (0.161) and  $g$   
 272 (0.521) were also typical for urban/suburban environments, suggesting a slightly stronger cooling potential of the aerosol  
 273 population compared to other sites.

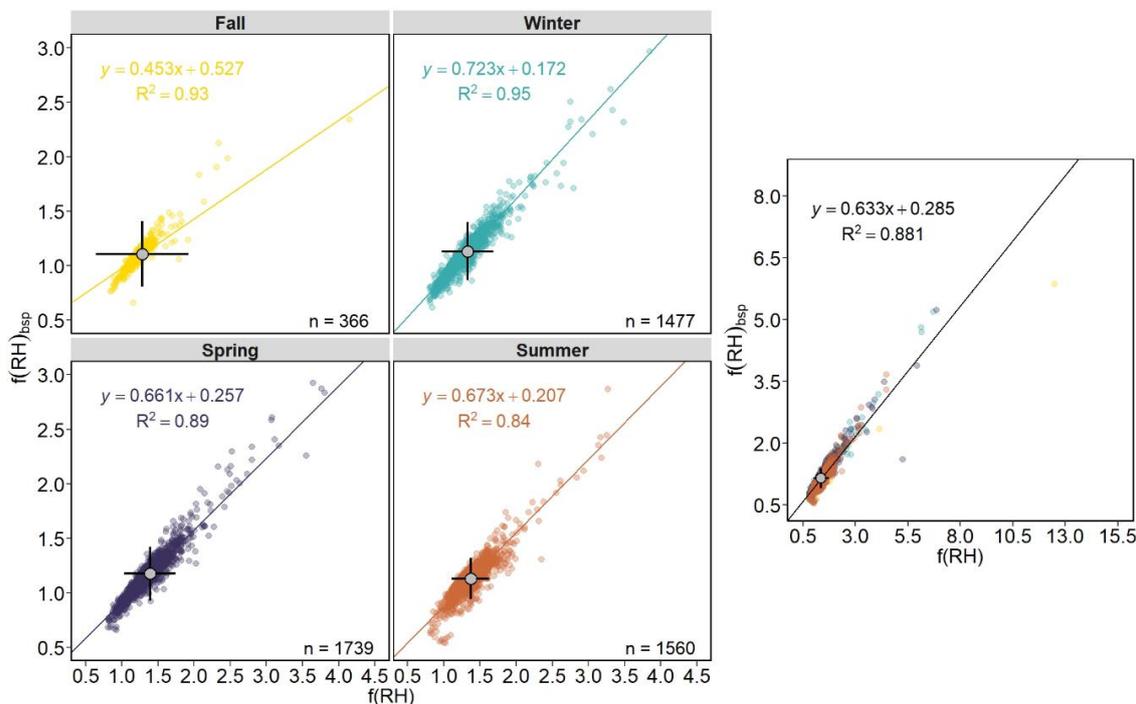
274 **Table 1: The statistics of light scattering properties in the  $PM_{10}$  fraction at different wavelengths. P25, P75, and P50 denote the 25<sup>th</sup>**  
 275 **and 75<sup>th</sup> percentiles and median, respectively. All variables except  $f(RH)$  and  $f(RH)_{bsp}$  were measured and calculated at  $RH < 40$  %, and all variables except  $\sigma_{sp}$  and  $\sigma_{bsp}$  are dimensionless.**

		Whole period				Fall	Winter	Spring	Summer
$\lambda$		P25	P50	P75	mean $\pm$ SD	P50			
$f(RH)$	450 nm	1.19	1.30	1.42	1.33 $\pm$ 0.34	1.20	1.27	1.34	1.33
	550 nm	1.20	1.32	1.44	1.35 $\pm$ 0.37	1.22	1.29	1.36	1.34
	700 nm	1.31	1.57	1.93	1.66 $\pm$ 0.54	1.46	1.54	1.61	1.63
$f(RH)_{bsp}$	450 nm	1.07	1.22	1.41	1.25 $\pm$ 0.32	1.20	1.20	1.25	1.20
	550 nm	1.04	1.12	1.20	1.14 $\pm$ 0.25	1.08	1.10	1.16	1.12
	700 nm	1.11	1.22	1.34	1.25 $\pm$ 0.32	1.15	1.20	1.27	1.22
$\sigma_{sp}$ ( $Mm^{-1}$ )	450 nm	21.81	40.31	80.29	60.99 $\pm$ 58.55	99.66	57.14	38.99	33.10
	550 nm	15.54	28.45	56.29	44.19 $\pm$ 43.50	73.22	40.84	27.99	22.96
	700 nm	10.89	19.27	36.76	29.93 $\pm$ 29.47	48.69	27.27	19.20	15.33
$\sigma_{bsp}$ ( $Mm^{-1}$ )	450 nm	3.37	6.05	10.33	8.14 $\pm$ 7.27	12.05	7.48	5.74	5.15
	550 nm	2.67	4.72	8.13	6.42 $\pm$ 5.70	9.54	5.86	4.56	3.97
	700 nm	2.29	4.08	7.13	5.62 $\pm$ 5.04	8.61	5.23	3.95	3.34
$SAE_{450-700}$		1.45	1.65	1.85	1.61 $\pm$ 0.34	1.56	1.57	1.70	1.81
$AAE_{470-660}$		1.29	1.46	1.61	1.45 $\pm$ 0.28	1.58	1.57	1.47	1.26
$\Delta SAE$		-0.091	0.008	0.194	0.063 $\pm$ 0.183	-0.122	-0.024	0.056	0.100
$SSA$	550 nm	0.744	0.805	0.851	0.794 $\pm$ 0.080	0.766	0.783	0.805	0.843
$b$		0.141	0.161	0.174	0.158 $\pm$ 0.027	0.134	0.151	0.163	0.171
$g$		0.492	0.521	0.562	0.528 $\pm$ 0.106	0.578	0.542	0.516	0.498

277 However, a relatively low median value of  $SSA_{550}$  (0.805) implies a substantial influence of absorbing aerosol species in  
 278 the aerosol population. The median  $AAE_{470-660}$  value of 1.46 suggests a relatively balanced contribution of black carbon (BC)  
 279 and brown carbon (BrC) to aerosol absorption, with a stronger influence of BC in summer ( $AAE_{470-660}$  of 1.26), likely due to  
 280 the increased presence of traffic emissions. In contrast, elevated AAE values in fall (1.58) and winter (1.57) suggest an  
 281 increased contribution of BrC from biomass burning, possibly related to residential heating.

### 3.2. $f(\text{RH})$ and $f(\text{RH})_{\text{bsp}}$

The overall low enhancement factors  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  of 1.32 and 1.12 suggest the influence of low-hygroscopic carbonaceous aerosol species from local combustion, traffic sources, and their aged derivatives. This result is consistent with the lower range of  $f(\text{RH})$  values observed at urban and suburban sites, for example, average values ranging between 1.32 and 1.74 in a suburban area of Beijing in autumn (Ren et al., 2021), 1.30 near Manacapuru city in Brazil, a site influenced by industrial activities with soot, high-sulfur oil emissions, and biomass burning (Burgos et al., 2019), or  $1.5 \pm 0.2$  in winter in Granada (Titos et al., 2014).



**Figure 4: The weighted bivariate fit of  $f(\text{RH})_{\text{bsp}}$  vs the  $f(\text{RH})$  at  $\lambda = 550$  nm for individual seasons (left). The grey point represents the overall  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  mean value with error bars. The right plot demonstrates the weighted bivariate fit for the whole dataset.**

The  $f(\text{RH})_{\text{bsp}}$  mimicked the behavior of the  $f(\text{RH})$  during the whole year, with the highest correlation in winter (Fig. 4). Compared to Titos et al. (2021), our mean  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  values (gray point in Fig. 4) fall into the low enhancement category of urban sites. Despite the strong linear relationship,  $f(\text{RH})_{\text{bsp}}$  does not precisely mirror  $f(\text{RH})$  and slightly varies mainly between colder (fall and winter) and warmer (spring and summer) seasons. As aerosol ERF depends on the hemispheric backscattering ratio  $b$ , models relying solely on assumed  $f(\text{RH})$  can lead to inaccurate outcomes (Haywood and Shine, 1995; Hegg et al., 1996; Titos et al., 2021). Such regressions can be beneficial given the scarcity of  $f(\text{RH})_{\text{bsp}}$  measurements for modeling enhancement for specific site types. Titos et al. (2021) performed a fit across several environments (Arctic, marine, rural, and urban) and found the fit equation of  $y = 0.55x + 0.32$ ,  $R^2=0.69$  for the Shouxian rural site in China. Parameters retrieved from our weighted bivariate fit of  $f(\text{RH})_{\text{bsp}}$  vs the  $f(\text{RH})$  can be found in Table S.3. These regression parameters describe the co-variation between  $f(\text{RH})_{\text{bsp}}$  and  $f(\text{RH})$ , whereas magnitude differences between the two enhancement factors are quantified below using median values.

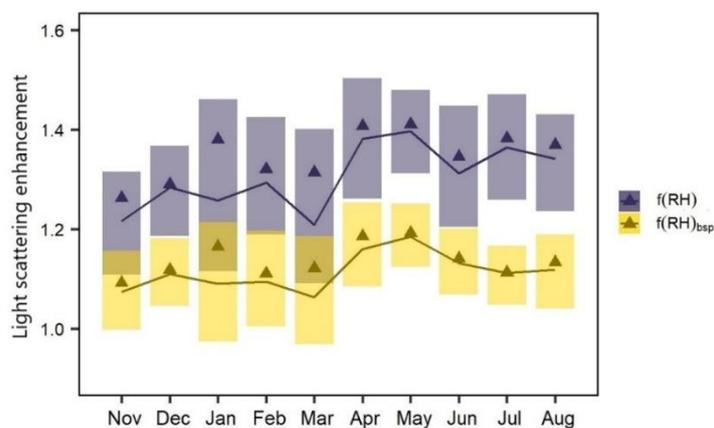
The  $f(\text{RH})$  depends on RH, the particle size, chemical composition, and light wavelength. Its spectral dependence is crucial for radiative forcing estimates (Fierz-Schmidhauser et al., 2010; Kiehl and Briegleb, 1993; Titos et al., 2021). At the studied site, the  $f(\text{RH})$  increases with wavelength in most cases and seasons except summer (frequency distributions centered around 0, Fig. S.3), aligning well with results from urban sites (Titos et al., 2021) but showing lower values compared to marine or Arctic environments. Occasionally, an opposite behavior with a decrease  $f(\text{RH})$  with increasing wavelength was observed, linked to dust episodes and particle size shift (Carrico et al., 2003; Fierz-Schmidhauser et al., 2010).

319 Based on Mie's theory, Hegg et al. (1996) proposed that  $f(\text{RH})_{\text{bsp}}$  should be approximately 25 % lower than  $f(\text{RH})$  for  
 320 typical atmospheric aerosols. Our observations support this, showing consistently smaller  $f(\text{RH})_{\text{bsp}}$  than  $f(\text{RH})$  across all  
 321 wavelengths. On average,  $f(\text{RH})_{\text{bsp}}$  was lower by 6 %, 15 %, and 22 % at 450, 550, and 700 nm, respectively, with the largest  
 322 differences observed in summer reaching 10 %, 16 %, and 25 %. The relative difference between  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  increases  
 323 with wavelength, suggesting a spectral sensitivity of backscattering to humidification, likely due to particle size and  
 324 composition effects on the angular distribution of scattered light. These findings highlight the need for wavelength- and season-  
 325 specific correction factors when using  $f(\text{RH})$  to estimate aerosol backscattering or when interpreting satellite data sensitive to  
 326 the backscattered light.  
 327 The probability density functions of  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  for different wavelengths can be found in Fig. S.4.

### 328 3.2.1. Seasonal variability

329 The lowest  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  values were observed in the fall, with a monthly minimum in November (1.22 and 1.07,  
 330 Fig. 5). Both peaked in spring, with a monthly maximum in May (1.40 and 1.19). The increase from autumn to spring was  
 331 interrupted in March when both  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  dropped to 1.22 and 1.08, probably due to March being a transitional  
 332 period between winter and spring in the Northern Hemisphere. Although this anomaly has not been fully understood yet,  
 333 CAMS reanalysis by Flemming et al. (2017) also identified irregular springtime atmospheric patterns over the Northern  
 334 Hemisphere, while Suchánková et al. (2025) observed a steep increase in dry  $\sigma_{\text{sp}}$  and  $\sigma_{\text{bsp}}$ , and ultrafine and fine particle number  
 335 concentration at an urban site in France. After peaking in May, both  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  gradually decreased towards August  
 336 (1.34 and 1.12). The seasonal variation is consistent with results reported at the urban site in Granada (Titos et al., 2014).

337 Lower values of  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  in fall and winter suggest the prevalence of low-hygroscopic aerosol species,  
 338 such as carbonaceous particles from combustion and traffic sources. In summer and spring, the enhancement is influenced by  
 339 hygroscopic SOA. Although  $\text{SSA}_{550}$  peaks in summer, likely due to highly hygroscopic secondary organic and inorganic  
 340 aerosols (sulfates and nitrates), the strongest correlation between  $f(\text{RH})/f(\text{RH})_{\text{bsp}}$  and  $\text{SSA}_{550}$  is in spring ( $R = 0.32/0.26$ ).  
 341 Sulfates and nitrates amplify aerosol hygroscopicity more effectively than organic matter, especially compared to the less  
 342 hygroscopic organic species prominent in summer. This seasonal contrast highlights the key role of inorganic species like  
 343 sulfate and nitrate in controlling aerosol optical properties through their impact on hygroscopic growth. During spring, the  
 344 aerosol composition is relatively homogeneous, dominated by aged, regionally transported aerosols with intermediate  
 345 hygroscopicity, resulting in stronger and more predictable correlations between enhancement factors and  $\text{SSA}_{550}$ . While both  
 346 spring and summer can include biogenic organics and aged urban particles, the chemical composition in summer is more  
 347 variable, leading to less consistent hygroscopic behavior and the weakest seasonal correlations with  $\text{SSA}_{550}$  ( $R = 0.16$  for both  
 348  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$ ).



355 **Figure 5: The monthly variation of the  $f(\text{RH})$  and the  $f(\text{RH})_{\text{bsp}}$  measured at 550 nm. The lines with shaded areas depict the median**  
 356 **with the interquartile range, and the triangles represent mean values.**

357 The temporal variation of  $f(\text{RH})$  did not show any consistent connection to  $\text{SAE}_{450-700}$  (Fig. S.5a). The  $\text{SAE}_{450-700}$  values  
358 rise from colder months to warmer months, suggesting the presence of smaller particles from the NPF or the traffic. Compared  
359 to winter, the enhanced mixing and higher dilution due to the higher planetary boundary layer height (PBLH) in spring and  
360 summer also prevent particle growth due to condensation. This statement was supported by the analysis of particle number  
361 size distribution (PNSD), with  $D_p$  below 200 nm primarily present in the photooxidatively active time of the year. At the same  
362 time, larger particles occurred mainly in colder seasons (Fig. S.6). However, the nephelometer does not precisely measure the  
363 ultrafine particle sizes due to its geometry and principle of operation, possibly distorting the direct relationship between the  
364 light scattering enhancement and PNSD.

365 Our results suggest that variations in aerosol chemical composition may play a more important role in modulating light  
366 scattering enhancement than particle size alone. The temporal evolution of  $f(\text{RH})$  and the ratio  $f(\text{RH})_{\text{bsp}}/f(\text{RH})$ , color-coded by  
367 dry  $\text{SAE}_{450-700}$  (Fig. S.5a,b), shows pronounced seasonal variability in particle size, while only weak co-variability of  $\text{SAE}_{450-}$   
368  $_{700}$  with increasing hygroscopic enhancement. Higher  $\text{SAE}_{450-700}$  values, predominantly observed in spring and summer,  
369 indicate a dominance of fine particles, consistent with an enhanced influence of NPF, whereas the remaining seasons reflect a  
370 mixture of fine and coarser particles typical of urban aerosol. Although smaller particles generally exhibit higher dry  
371 backscattering fractions, hygroscopic growth can modify particle size and the scattering phase function, leading to a  
372 comparatively weaker enhancement of backscattering than total scattering (Fig. S.5b). This interpretation is further supported  
373 by seasonal changes in particle number size distributions (Fig. S.6), which indicate shifts in dominant size modes, reflecting  
374 changes in aerosol sources over the year. It should be noted that this interpretation is based on indirect proxies for  
375 aerosol composition and size, as no direct chemical analysis is available, and particle size distributions are limited  
376 to  $\text{PM}_{10}$ , while hygroscopic scattering measurements represent  $\text{PM}_{10}$ .

### 377 3.3. Light scattering enhancement vs other aerosol-intensive optical properties

378 Understanding the relationship between light scattering enhancement and other aerosol optical properties can help improve  
379 the characterization of aerosol types and their radiative impacts.

380  $\text{SAE}_{450-700} > 1$  mirrored the frequency distribution of both  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  (Fig. 6). A slight shift towards more hygroscopic  
381 behavior was observed for  $\text{SAE}_{450-700} < 1$  in  $f(\text{RH})_{\text{bsp}}$ . In contrast,  $\text{SSA}_{550} < 0.8$  exhibits a slight shift to the left for both  $f(\text{RH})$   
382 and  $f(\text{RH})_{\text{bsp}}$ , indicating an overall significant decreasing effect of absorbing aerosol species on aerosol light scattering  
383 enhancement, consistent with the finding of Titos et al. (2014).

384 In winter and spring, particles with  $\text{SAE}_{450-700} < 1$  showed slightly more hygroscopic behavior both for  $f(\text{RH})$  and  
385  $f(\text{RH})_{\text{bsp}}$ , possibly due to the internal mixtures of sulfates and nitrates or the presence of mineral dust. Particles characterized  
386 with  $\text{SSA}_{550} < 0.8$  consistently showed lower  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  values. In spring and summer, the frequency distribution of  
387  $\text{SSA}_{550} > 0.8$  mimicked the whole dataset distribution, implying a higher share of light-scattering aerosols (Fig. S.7).

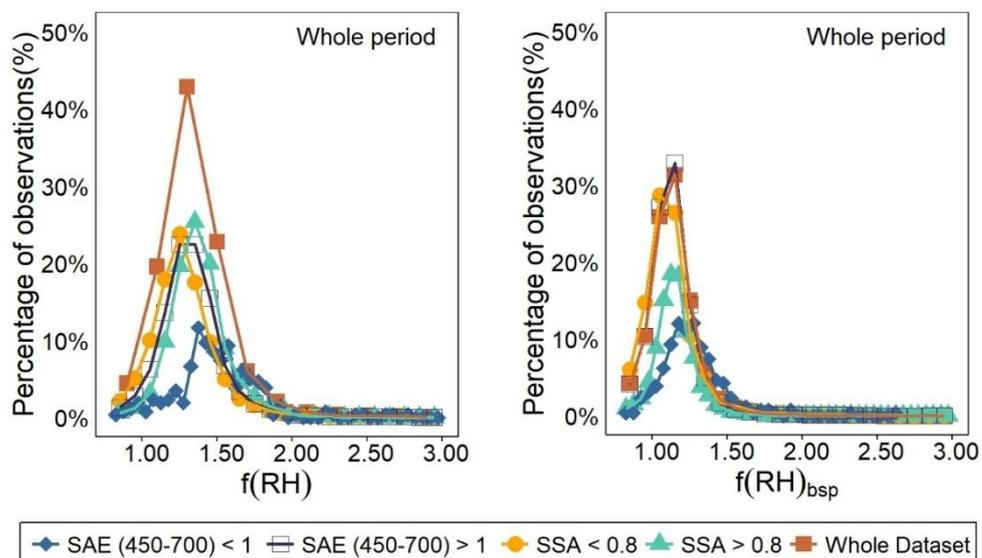
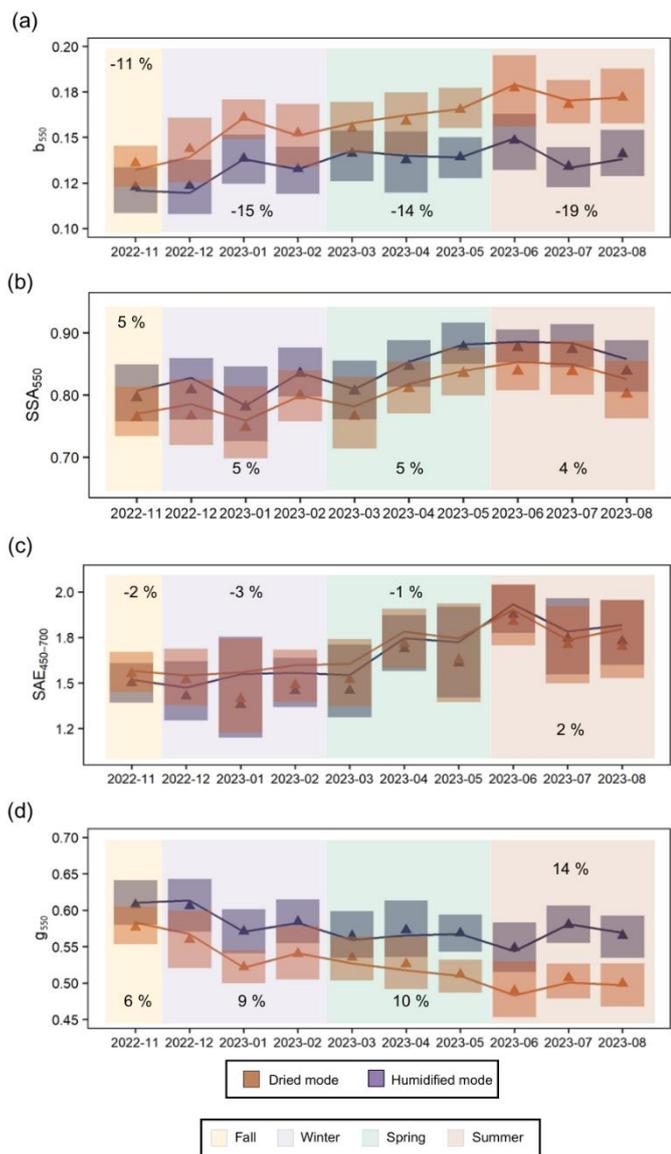


Figure 6: Frequency distribution of  $f(RH)$  (left) and  $f(RH)_{bsp}$  (right) calculated at 550 nm during the whole measurement campaign. The distributions are color and symbol-coded based on  $SAE_{450-700}$  and  $SSA_{550}$  values.

Dry  $SSA_{550}$  was lower for less hygroscopic particles ( $f(RH) < 1.5$ ) than for more hygroscopic ones ( $f(RH) > 1.5$ ), 0.708 vs 0.742 (Fig. S.8a), confirming the adverse effect of absorbing components on hygroscopicity. Across all seasons, the  $SSA$  increased with  $RH$  (Fig. 7b), while  $b_{550}$  showed the opposite behavior (Fig. 7a), as expected, given their inverse relationship. This relationship was more pronounced under humidified conditions than dried ones (Fig. S.8b) (Burgos et al., 2020; Titos et al., 2021). As particles take up water and grow, the total scattering increases with a predominance of the forward light scattering over backscattering.

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421 **Figure 7: The monthly variation of  $b_{550}$  (a),  $SSA_{550}$  (b),  $SAE_{450-700}$  (c), and  $g_{550}$  (d) at dried and humidified conditions in individual**  
 422 **seasons (colored background). A percentage value in each season shows a relative change between humidified and dried median**  
 423 **values for the respective season. The lines with shaded areas depict the median and the interquartile range, and the triangles**  
 424 **represent respective mean values.**

425 The asymmetry factor  $g_{550}$  also increased with RH, confirming stronger forward than backward scattering with enhanced  
 426 RH (Fig. 7d). The  $SAE_{450-700}$  decreased with increasing RH, indicating particle size growth in all seasons except summer (Fig.  
 427 7c). During summer, an atypical behavior of the  $SAE_{450-700}$  was observed, with dry particles appearing optically larger (lower  
 428 SAE) than the humidified ones. This is likely linked to the dominance of BC-rich aerosols, which are weakly hygroscopic and  
 429 maintain large agglomerated structures in the dry state. Such structures may compact upon humidification, reducing optical  
 430 growth and increasing SAE. This behavior contrasts with highly hygroscopic sulfate- or nitrate-rich aerosol regimes, where  
 431 humidification leads to particle growth and lower SAE values.

432 Comparing our site with those reported by Titos et al. (2021) we found the lowest  $f(RH)$ , similar to urban (e.g.,  
 433 Manacapuru, Brazil, and Nainital, India), desert (Niamey, Niger), and some rural (e.g., Hyytiälä, Finland) environments.  
 434 Interestingly, the  $SSA_{550}$  at our site was the lowest, indicating a strong presence of absorbing aerosols such as black carbon or  
 435 other combustion-related species (Fig. S.5c). This confirms the role of local or regional combustion sources in shaping aerosol  
 436 optical properties.

### 3.4. The estimation of chemical composition from optical properties

Due to the lack of direct measurement of chemical composition at the site, we used the approach introduced by Cappa et al. (2016), using  $AAE_{520-660}$ ,  $SAE_{450-550}$ , and  $SSA_{550}$  to estimate the aerosol chemical composition. Aerosol at the station was found mainly in the fine fraction, dominated by BC in a mixture with BrC (Fig. 8), in agreement with the low  $f(RH)$  and  $f(RH)_{bsp}$  at the site. The site was also influenced by BC and BrC in a mixture with dust, probably coming from the road dust, although unexamined Saharan dust episodes may have contributed. These mixtures included large, poorly absorbing coated particles and locally emitted sulfates and nitrates in the fine size mode, which highly scatter but weakly absorb the incoming light.

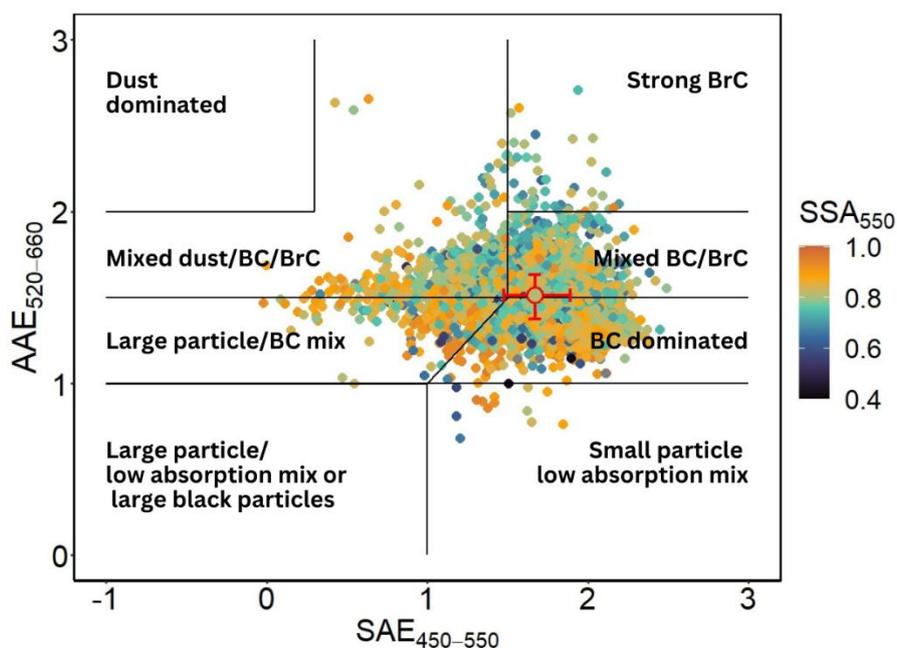
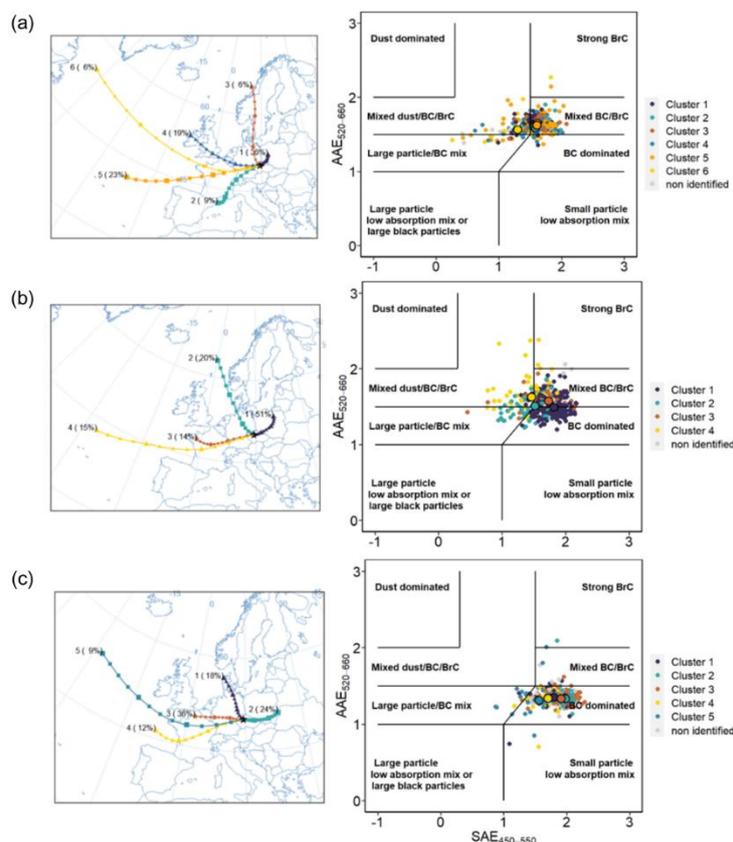


Figure 8:  $AAE_{520-660}$  vs.  $SAE_{450-550}$  hourly means from the whole campaign, color-coded with  $SSA_{550}$  overlaid with the aerosol characterization matrix adopted from Cappa et al. (2016) and Cazorla et al. (2013). The red circle with red error bars estimated the median  $AAE_{520-660}$ – $SAE_{450-550}$  point with interquartile ranges.

$SSA_{550}$  values were equally distributed among the plot regions, except for the highest values in the aerosol mixture of sea salt, dust, and low-absorbing coated particles with BC and BrC. Such aerosol was typically connected with air masses coming from Poland and Eastern Europe (Fig. 9). High concentrations of the mixture with BrC were observed in fall, winter, and spring, predominantly due to biomass burning. Consequently, the median  $AAE_{520-660}$  vs.  $SAE_{450-550}$  point fell into the "Mixed/BC/BrC" plot region. In contrast, the BC aerosol mixture started to dominate in spring and summer (Fig. S.9).

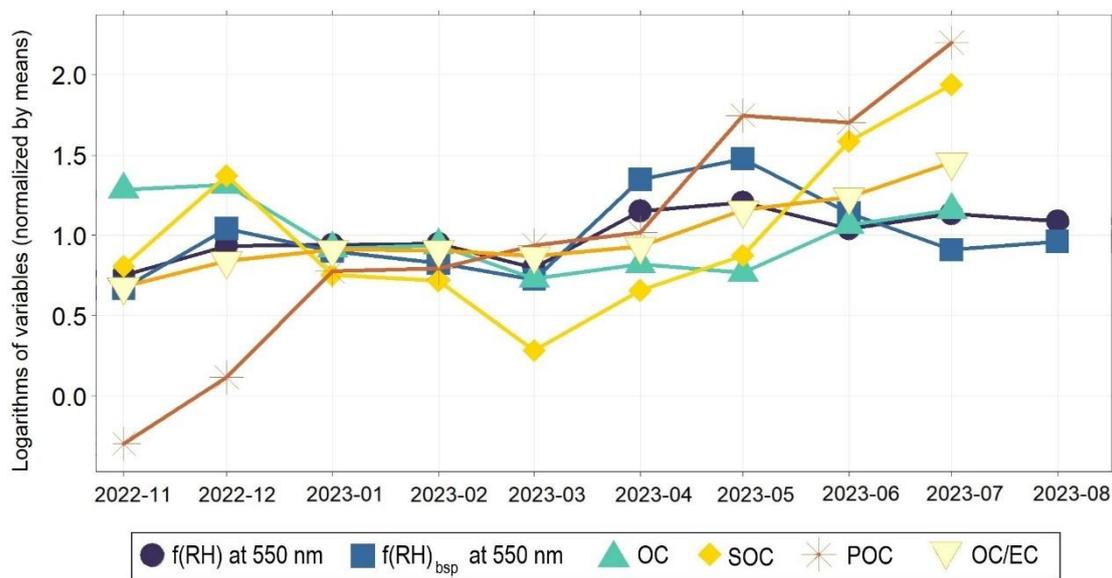
Although multiple wintertime clusters originated from the marine regions, only Cluster 6 (Fig. 9a) shifted towards dust/large low-absorbing particle aerosol mixture, indicating the influence of fossil fuel/biomass burning sources during this season. In spring, the air masses of Clusters 2 and 4 originated from the Norwegian Sea and Atlantic Ocean (Fig. 9b) and featured hygroscopic sea salt mixtures. In summer, the BC-dominated aerosol prevailed (Fig. 9c); however, marine clusters (Clusters 1, 4, and 5) shifted towards larger aerosol sizes, suggesting aerosol-aged mixtures influencing the ability to scatter and absorb radiation.



460 **Figure 9: The 72-h air mass back trajectories paired with AAE520-660 vs SAE450-550 for winter (a), spring (b), and summer (c).**  
 461 **The plots are color-coded by trajectory cluster number, with cluster medians indicated by black circles and the percentage**  
 462 **contribution of each cluster provided. The fall was not taken into consideration due to the limited data.**

463 Consistent with previous hygroscopic studies, aerosol hygroscopic changes are often negatively correlated with the  
 464 organic mass fraction, reflecting the low hygroscopicity of ambient organic aerosol compared to strongly hygroscopic  
 465 inorganic salts (mainly sulphates and nitrates) (Andrews et al., 2021; Kang et al., 2025; Massoli et al., 2009; Pöhlker et al.,  
 466 2023). We note that direct measurements of inorganic aerosol components were not available at the Suchdol site, and therefore  
 467 their contributions to  $f(\text{RH})$  variability cannot be quantified in this study. Nevertheless, a substantial fraction of the variability  
 468 in  $f(\text{RH})$  is likely controlled by variations in the inorganic fraction, and correlations between  $f(\text{RH})$  and organic metrics can  
 469 partly reflect this co-variability. Separating OC into primary (POC) and secondary (SOC) fractions allows investigation of  
 470 differences in effective hygroscopicity within the organic aerosol itself. To further explore variability in optical hygroscopicity  
 471 beyond total organic mass, semi-online OC/EC measurements were used to calculate the concentrations of secondary (SOC)  
 472 and primary (POC) organic carbon, following the method of Mbengue et al. (2021). The concentrations of OC, POC, SOC,  
 473 and the ratio of organic and elemental carbon (OC/EC) remained stable during winter and increased in spring, mirroring the  
 474 behavior of  $f(\text{RH})$  until May (Fig. 10). However, carbonaceous components rose from late spring into summer while mainly  
 475  $f(\text{RH})_{\text{bsp}}$  decreased, suggesting a seasonal decoupling of SOC and aerosol hygroscopicity. This may be due to the higher  
 476 fraction of POC (Liu and Wang, 2010), the formation of less hygroscopic SOA from biogenic precursors (Huang et al., 2019),  
 477 and the evaporation of semi-volatile organics at higher temperatures, reducing their contribution to water uptake and  $f(\text{RH})$   
 478 (Thomsen et al., 2024).

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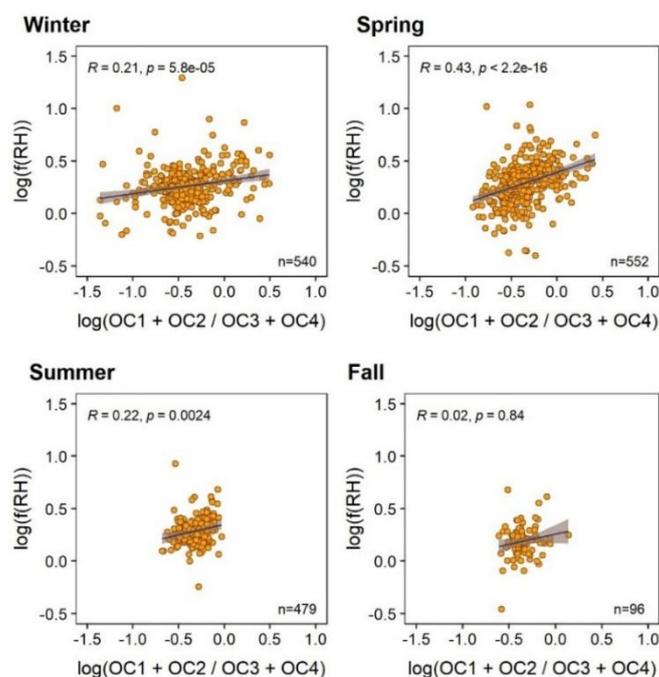
**Figure 10: Temporal variation of logarithmic median values of f(RH) and f(RH)<sub>bsp</sub>, OC, SOC, POC, and OC/EC. Each series was divided by its mean value to compensate for different numeric scales.**

Elemental carbon (EC) showed negative Spearman correlations with f(RH) and f(RH)<sub>bsp</sub> ( $R = -0.33$  and  $-0.17$ ), as expected. OC showed a moderate negative correlation with f(RH), yet the OC/EC ratio correlated positively with f(RH) and f(RH)<sub>bsp</sub>, especially during spring ( $R = 0.31$ ). This indicates that the relative abundance of organic carbon, rather than its absolute concentration, enhances hygroscopicity (Table S.4). The SOC/OC ratio seasonal behavior followed the OC/EC seasonality, emphasizing the role of SOA in aerosol hygroscopic growth.

The four OC subfractions distinguished by the thermo-optical method further clarified this behavior. Surprisingly, the sum of more volatile OC1 and OC2 positively correlates with f(RH). In contrast, the sum of less volatile OC3 and OC4 showed negative correlations, implying higher hygroscopicity of fresher, more volatile OC subfractions (Fig. 11). This aligns with field studies of “smoke” particles by Chan et al. (2005), showing that levoglucosan from biomass burning or urban pollution can “age” into simpler dicarboxylic acids, such as succinic acid, which is less hygroscopic than the original substance, causing aged biomass burning aerosols to be less hygroscopic than the fresh ones. However, the hygroscopicity of the mentioned organic compounds highly depends on their mixing state and the additional components in the mixture (Maskey et al., 2014).

A key factor influencing this trend may be Humic-Like Substances (HULIS), typically associated with OC3 and OC4. HULIS are low in volatility and hygroscopicity (Kristensen et al., 2012) and are primarily emitted from biomass burning, residential heating, and traffic. Their presence may suppress water uptake and light scattering (Dinar et al., 2008). Urban HULIS tend to absorb light more and decrease SSA than rural sources (Tang et al., 2020). HULIS often exist in the glassy or semi-solid state, preventing diffusion of water molecules in their structure and decreasing their hygroscopicity (Koop et al., 2011), and further, aging can enhance viscosity via oligomerization, reducing hygroscopicity even more (Song et al., 2016).

The highest correlation between f(RH), f(RH)<sub>bsp</sub>, and (OC1 + OC2)/(OC3 + OC4) ratio was found in spring ( $R=0.43$  and  $0.28$ ) despite stronger photochemical activity during summer (Fig. 11). This counterintuitive result suggests that spring conditions preserve more water-uptake-efficient OC1 and OC2 in the particulate phase, while intense summer photochemistry and higher temperatures drive oxidation and oligomerization of organic compounds, transforming initially hygroscopic semi-volatile organics (OC1+OC2) into more aged, less hygroscopic fractions (OC3+OC4), and promote volatile organics evaporation, reducing the relative contribution of fresh OC to the particulate phase.



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517 **Figure 11: Seasonal Spearman correlations between the  $f(\text{RH})$  and the log-transformed ratio of more volatile OC fractions**  
 518 **(OC1+OC2) and less volatile OC fractions (OC3+OC4). The values in the bottom right corner indicate the number of observations,**  
 519 **and the values in the upper left corner indicate the Spearman correlation coefficient  $R$  and the respective  $p$ -values.**

520 **4. Summary and conclusions**

521 This work presented a cost-effective approach to investigate aerosol hygroscopicity using a single-nephelometer set-up  
 522 with an automatically controlled switching valve alternating between humidified and dried sample branches. This design  
 523 reduced uncertainties associated with dual-instrument configurations while allowing for investigation of light backscattering  
 524 changes with elevated RH. After testing and calibrations, the system was installed at the suburban site, Suchdol in Prague,  
 525 Czech Republic, from November 2022 to August 2023 to fill the knowledge gap related to the optical hygroscopic behavior of  
 526 aerosol at European urban/suburban sites, particularly regarding light scattering under increased relative humidity (RH).

527 The light total scattering enhancement factor  $f(\text{RH})$  and the light backscattering enhancement factor  $f(\text{RH})_{\text{bsp}}$  were derived  
 528 from the humidified and dried measurement of the light total scattering ( $\sigma_{\text{sp}}$ ) and backscattering ( $\sigma_{\text{bsp}}$ ) coefficients at 450, 550,  
 529 and 700 nm. These enhancement factors were analyzed in relation to climate-relevant aerosol optical properties, including the  
 530 Scattering/Absorption Ångström Exponent SAE/AE, hemispheric backscattering ratio  $b$ , asymmetry factor  $g$ , and Single  
 531 Scattering Albedo SSA, as well as estimated chemical composition, particle number size distributions (PNSD), meteorological  
 532 parameters, and back trajectory analysis.

533 The measured  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  at 550 nm (1.32 and 1.12) were among the lowest reported in comparable studies,  
 534 together with one of the lowest  $\text{SSA}_{550}$  values. This indicates a dominance of low-hygroscopic aerosol species originating from  
 535 local combustion and traffic emissions. Seasonal and wavelength-dependent differences in the relationship between  $f(\text{RH})_{\text{bsp}}$   
 536 and  $f(\text{RH})$  highlight the need for aerosol-type-specific parametrizations in radiative models. Peaks in both enhancement factors  
 537 were detected in spring, coinciding with increased contributions of more hygroscopic aerosol components and enhanced  
 538 photochemical activity. Increasing  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  were associated with decreasing hemispheric backscattering ratios and  
 539 increasing asymmetry factors and SSA, indicating enhanced total scattering and stronger forward scattering due to hygroscopic  
 540 growth. In contrast, during summer,  $\text{SAE}_{450-700}$  increased with RH, suggesting particle shrinkage, likely linked to the  
 541 dominance of low-hygroscopic black carbon (BC) emissions, which is prone to structural compaction upon humidification.

542 Chemical composition was found to exert a stronger control on light scattering enhancement than particle number  
 543 size distribution. BC-dominated aerosol prevailed in summer, while other seasons were characterized by mixed aerosol

544 populations containing BC, brown carbon (BrC), and episodic dust or marine influences. Both  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$  showed  
545 positive correlations with OC/EC and SOC/OC ratios. More volatile organic fractions exhibited stronger associations with the  
546 light scattering enhancement than less volatile fractions, particularly in the spring, underscoring the important role of secondary  
547 organic carbon in modulating aerosol hygroscopicity in carbon-dominated urban environments.

548 In summary, the single-nephelometer system proved suitable for long-term ambient aerosol characterization. The  
549 main limitations of the approach are reduced time resolution and the absence of simultaneous dry and wet measurements  
550 required for detailed humidogram analysis. Nevertheless, this study contributes valuable observations to the global dataset by  
551 characterizing environments where low  $f(\text{RH})$ , low SSA, and relatively high SAE coexist—conditions that remain  
552 underrepresented in current aerosol–climate literature.

### 553 **Authors contribution**

554 LS, JO, PR and VZ designed the methodology of the work and created the study conceptualization. Additionally, LS was  
555 responsible for data curation, formal analysis, investigation, validation, software, visualization, and writing of the original  
556 draft. JO was responsible for software, resources, investigation, and writing — review & editing. NZ contributed to data  
557 curation, formal analysis and writing — review & editing. PR was additionally responsible for software. PV was responsible  
558 for data curation and writing — review & editing. RP was responsible for project administration, funding and administration and  
559 writing — review & editing. IH contributed to supervision. VZ was responsible for project administration, supervision,  
560 resources and writing — review & editing.

### 561 **Data availability**

562 Datasets including  $f(\text{RH})$  and  $f(\text{RH})_{\text{bsp}}$ , original relative humidity and temperature variation within humidified single-  
563 nephelometer set-up system, concentration of carbonaceous aerosol species, particle size number concentration,  
564 meteorological parameters, dry and humidified aerosol optical properties, NPF categorization, and air mass back-trajectory  
565 cluster identification are available at Suchánková, Lenka (2025), “Humidified single-nephelometer set-up system datasets”,  
566 Mendeley Data, V1, doi: 10.17632/8ds98t2f3x.1.

### 567 **Competing interests**

568 The authors declare that they have no competing interests.

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### 577 **References**

578 Alduchov, O. A. and Eskridge, R. E.: Magnus-Tetens formula, Asheville, 21 pp.,  
579 <https://doi.org/https://doi.org/10.2172/548871>, 1997.

580 Anderson, T. L. and Ogren, J. A.: Determining Aerosol Radiative Properties Using the TSI 3563 Integrating Nephelometer,

581 Aerosol Sci. Technol., 29, 57–69, <https://doi.org/10.1080/02786829808965551>, 1998.

582 Anderson, T. L., Covert, D. S., Marshall, S., Laucks, M. L., Charlson, R. J., Waggoner, A. P., Ogren, J. A., Caldow, R., Holm,  
583 R. L., Quant, F. R., Sem, G. J., Wiendensholer, A., Ahlquist, N. A., and Bates, T. S.: Performance Characteristics of a High-  
584 Sensitivity, Three-Wavelength, Total Scatter/Backscatter Nephelometer, *Cover J. Atmos. Ocean. Technol. J. Atmos. Ocean.*  
585 *Technol.*, 13, 967–986, [https://doi.org/https://doi.org/10.1175/1520-0426\(1996\)013<0967:PCOAHS>2.0.CO;2](https://doi.org/https://doi.org/10.1175/1520-0426(1996)013<0967:PCOAHS>2.0.CO;2), 1996.

586 Andrews, E., Sheridan, P. J., Fiebig, M., McComiskey, A., Ogren, J. A., Arnott, P., Covert, D., Elleman, R., Gasparini, R.,  
587 Collins, D., Jonsson, H., Schmid, B., and Wang, J.: Comparison of methods for deriving aerosol asymmetry parameter, *J.*  
588 *Geophys. Res. Atmos.*, 111, D05S04, <https://doi.org/10.1029/2004JD005734>, 2006.

589 Andrews, E., Zieger, P., Titos, G., and Burgos, M.: Evaluation and improvement of the parameterization of aerosol  
590 hygroscopicity in global climate models using in-situ surface measurements (Final Report), <https://doi.org/10.2172/1706478>,  
591 2021.

592 Aswini, A. R., Hegde, P., Nair, P. R., and Aryasree, S.: Seasonal changes in carbonaceous aerosols over a tropical coastal  
593 location in response to meteorological processes, *Sci. Total Environ.*, 656, 1261–1279,  
594 <https://doi.org/10.1016/J.SCITOTENV.2018.11.366>, 2019.

595 Bauer, J. J., Yu, X.-Y., Cary, R., Laulainen, N., and Berkowitz, C.: Characterization of the sunset semi-continuous carbon  
596 aerosol analyzer., *J. Air Waste Manag. Assoc.*, 59, 826–833, <https://doi.org/10.3155/1047-3289.59.7.826>, 2009.

597 Boucher, O.: Atmospheric Aerosols, in: *Atmospheric Aerosols*, Springer Netherlands, Dordrecht, 9–24,  
598 [https://doi.org/10.1007/978-94-017-9649-1\\_2](https://doi.org/10.1007/978-94-017-9649-1_2), 2015.

599 Burgos, M. A., Andrews, E., Titos, G., Alados-Arboledas, L., Baltensperger, U., Day, D., Jefferson, A., Kalivitis, N.,  
600 Mihalopoulos, N., Sherman, J., Sun, J., Weingartner, E., and Zieger, P.: A global view on the effect of water uptake on aerosol  
601 particle light scattering, *Sci. Data*, 6, 1–19, <https://doi.org/10.1038/s41597-019-0158-7>, 2019.

602 Burgos, M. A., Andrews, E., Titos, G., Benedetti, A., Bian, H., Buchard, V., Curci, G., Kipling, Z., Kirkevåg, A., Kokkola,  
603 H., Laakso, A., Letertre-Danczak, J., T. Lund, M., Matsui, H., Myhre, G., Randles, C., Schulz, M., Van Noije, T., Zhang, K.,  
604 Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Sun, J., Weingartner, E., and Zieger, P.: A global model-  
605 measurement evaluation of particle light scattering coefficients at elevated relative humidity, *Atmos. Chem. Phys.*, 20, 10231–  
606 10258, <https://doi.org/10.5194/ACP-20-10231-2020>, 2020.

607 Cappa, C. D., Kolesar, K. R., Zhang, X., Atkinson, D. B., Pekour, M. S., Zaveri, R. A., Zelenyuk, A., and Zhang, Q.:  
608 Understanding the optical properties of ambient sub- and supermicron particulate matter: Results from the CARES 2010 field  
609 study in northern California, *Atmos. Chem. Phys.*, 16, 6511–6535, <https://doi.org/10.5194/acp-16-6511-2016>, 2016.

610 Carrico, C. M., Kus, P., Rood, M. J., Quinn, P. K., Bates, T. S., Carrico, C. :, Kus, P., Rood, M. J., Quinn, P. K., and Bates, T.  
611 S.: Mixtures of pollution, dust, sea salt, and volcanic aerosol during ACE-Asia: Radiative properties as a function of relative  
612 humidity, *J. Geophys. Res. Atmos.*, 108, 8650, <https://doi.org/10.1029/2003JD003405>, 2003.

613 Carslaw, K. S., Gordon, H., Hamilton, D. S., Johnson, J. S., Regayre, L. A., Yoshioka, M., and Pringle, K. J.: Aerosols in the  
614 Pre-industrial Atmosphere, *Curr. Clim. Chang. Reports* 2017 31, 3, 1–15, <https://doi.org/10.1007/S40641-017-0061-2>, 2017.

615 Cavalli, F., Viana, M., Yttri, K. E., Genberg, J., and Putaud, J. P.: Toward a standardized thermal-optical protocol for measuring  
616 atmospheric organic and elemental carbon: The EUSAAR protocol, *Atmos. Meas. Tech.*, 3, 79–89,  
617 <https://doi.org/10.5194/amtd-2-2321-2009>, 2010.

618 Cazorla, A., Bahadur, R., Suski, K. J., Cahill, J. F., Chand, D., Schmid, B., Ramanathan, V., and Prather, K. A.: Relating  
619 aerosol absorption due to soot, organic carbon, and dust to emission sources determined from in-situ chemical measurements,  
620 *Atmos. Chem. Phys.*, 13, 9337–9350, <https://doi.org/10.5194/acp-13-9337-2013>, 2013.

621 Statistický atlas - ČSÚ:  
622 <https://geodata.csu.gov.cz/as/atlas/?xmax=2194777.485518976&ymin=6135884.7339584185&wkid=102100>, last access: 13 December 2024.  
623

624 Chan, M. N., Choi, M. Y., Ng, N. L., and Chan, C. K.: Hygroscopicity of water-soluble organic compounds in atmospheric  
625 aerosols: Amino acids and biomass burning derived organic species, *Environ. Sci. Technol.*, 39, 1555–1562,  
626 <https://doi.org/10.1021/ES049584L>, 2005.

627 Clarke, A. and Kapustin, V.: Hemispheric aerosol vertical profiles: Anthropogenic impacts on optical depth and cloud nuclei,  
628 *Science*, 329, 1488–1492, [https://doi.org/10.1126/SCIENCE.1188838/SUPPL\\_FILE/CLARKE.SOM.PDF](https://doi.org/10.1126/SCIENCE.1188838/SUPPL_FILE/CLARKE.SOM.PDF), 2010.

629 Covert, D. S., Charlson, R. J., and Ahlquist, N. C.: A Study of the Relationship of Chemical Composition and Humidity to  
630 Light Scattering by Aerosols, *J. Appl. Meteorol.*, 11, 968–976, [https://doi.org/10.1175/1520-0450\(1972\)011<0968:asotro>2.0.co;2](https://doi.org/10.1175/1520-0450(1972)011<0968:asotro>2.0.co;2), 1972.  
631

632 Dinar, E., Abo Riziq, A., Spindler, C., Erlick, C., Kiss, G., and Rudich, Y.: The complex refractive index of atmospheric and  
633 model humic-like substances (HULIS) retrieved by a cavity ring down aerosol spectrometer (CRD-AS), *Faraday Discuss.*,  
634 137, 279–295, <https://doi.org/10.1039/B703111D>, 2008.

635 Doherty, S. J.: A comparison and summary of aerosol optical properties as observed in situ from aircraft, ship, and land during  
636 ACE-Asia, *J. Geophys. Res.*, 110, D04201, <https://doi.org/10.1029/2004JD004964>, 2005.

637 Donateo, A., Lo Feudo, T., Marinoni, A., Calidonna, C. R., Contini, D., and Bonasoni, P.: Long-term observations of aerosol  
638 optical properties at three GAW regional sites in the Central Mediterranean, *Atmos. Res.*, 241, 104976,  
639 <https://doi.org/10.1016/j.atmosres.2020.104976>, 2020.

640 Draxler, R. R. and Hess, G. D.: An overview of the HYSPLIT\_4 modelling system for trajectories, dispersion and deposition,  
641 1998.

642 Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T.,  
643 Wiedensohler, A., and Hansen, A. D. A.: The “dual-spot” Aethalometer: An improved measurement of aerosol black carbon  
644 with real-time loading compensation, *Atmos. Meas. Tech.*, 8, 1965–1979, <https://doi.org/10.5194/AMT-8-1965-2015>, 2015.

645 Fierz-Schmidhauser, R., Zieger, P., Gysel, M., Kammermann, L., Decarlo, P. F., Baltensperger, U., and Weingartner, E.:  
646 Measured and predicted aerosol light scattering enhancement factors at the high alpine site Jungfraujoch, *Atmos. Chem. Phys.*,  
647 10, 2319–2333, 2010.

648 Flemming, J., Benedetti, A., Inness, A., Engelen, J. R., Jones, L., Huijnen, V., Remy, S., Parrington, M., Suttie, M., Bozzo, A.,  
649 Peuch, V. H., Akritidis, D., and Katragkou, E.: The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for  
650 2003-2015, *Atmos. Chem. Phys.*, 17, 1945–1983, <https://doi.org/10.5194/acp-17-1945-2017>, 2017.

651 Foster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J. L., Frame, D., Lunt, D. J., Mauritsen, T., Palmer, M. D.,  
652 Watanabe, M., Wild, M., and Zhang, H.: The Earth’s Energy Budget, Climate Feedbacks, and Climate Sensitivity. In *Climate  
653 Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the  
654 Intergovernmental Panel on Climate Change, Climate Change 2021 – The Physical Science Basis*, Cambridge University Press,  
655 923–1054 pp., <https://doi.org/10.1017/9781009157896.009>, 2023.

656 Haywood, J. M. and Shine, K. P.: The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation  
657 budget, *Geophys. Res. Lett.*, 22, 603–606, <https://doi.org/10.1029/95GL00075>, 1995.

658 Hegg, D. A., Covert, D. S., Rood, M. J., and Hobbs, P. V.: Measurements of aerosol optical properties in marine air, *J. Geophys.*  
659 *Res. Atmos.*, 101, 12893–12903, <https://doi.org/10.1029/96JD00751>, 1996.

660 Held, A., Zerrath, A., McKeon, U., Fehrenbach, T., Niessner, R., Plass-Dülmer, C., Kaminski, U., Berresheim, H., and Pöschl,  
661 U.: Aerosol size distributions measured in urban, rural and high-alpine air with an electrical low pressure impactor (ELPI),  
662 *Atmos. Environ.*, 42, 8502–8512, <https://doi.org/10.1016/j.atmosenv.2008.06.015>, 2008.

663 Huang, W., Saathoff, H., Shen, X., Ramisetty, R., Leisner, T., and Mohr, C.: Seasonal characteristics of organic aerosol  
664 chemical composition and volatility in Stuttgart, Germany, *Atmos. Chem. Phys.*, 19, 11687–11700,  
665 <https://doi.org/10.5194/ACP-19-11687-2019>, 2019.

666 IPCC: Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report  
667 of the Intergovernmental Panel on Climate Change [Masson-Delmotte, V., P. Zhai, A. Pirani, S. L. Connors, C. Péan, S.  
668 Berger, N. Caud, Y. Chen, edited by: Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S. L., Péan, C., Berger, S., Caus, N.,  
669 Chen, Y., Goldfarb, L., Gomis, M. I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J. B. R., Maycock, T. K., Waterfield, T.,  
670 Yelekçi, O., Yu, R., and Zhou, B., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 3949  
671 pp., 2021.

672 Kahn, R. A., Andrews, E., Brock, C. A., Chin, M., Feingold, G., Gettelman, A., Levy, R. C., Murphy, D. M., Nenes, A., Pierce,  
673 J. R., Popp, T., Redemann, J., Sayer, A. M., da Silva, A. M., Sogacheva, L., and Stier, P.: Reducing Aerosol Forcing  
674 Uncertainty by Combining Models With Satellite and Within-The-Atmosphere Observations: A Three-Way Street, *Rev.*  
675 *Geophys.*, 61, <https://doi.org/10.1029/2022RG000796>, 2023.

676 Kang, H., Jung, C. H., Lee, B. Y., Krejci, R., Heslin-Rees, D., Aas, W., and Yoon, Y. J.: Aerosol hygroscopicity influenced  
677 by seasonal chemical composition variations in the Arctic region, *J. Aerosol Sci.*, 186, 106551,  
678 <https://doi.org/10.1016/j.jaerosci.2025.106551>, 2025.

679 Kiehl, J. T. and Briegleb, B. P.: The Relative Roles of Sulfate Aerosols and Greenhouse Gases in Climate Forcing, *Science*,  
680 260, 311–314, <https://doi.org/10.1126/SCIENCE.260.5106.311>, 1993.

681 Koop, T., Bookhold, J., Shiraiwa, M., and Pöschl, U.: Glass transition and phase state of organic compounds: dependency on  
682 molecular properties and implications for secondary organic aerosols in the atmosphere, *Phys. Chem. Chem. Phys.*, 13, 19238–  
683 19255, <https://doi.org/10.1039/C1CP22617G>, 2011.

684 Kristensen, T. B., Wex, H., Nekat, B., Njgaard, J. K., Van Pinxteren, D., Lowenthal, D. H., Mazzoleni, L. R., Dieckmann, K.,  
685 Bender Koch, C., Mentel, T. F., Herrmann, H., Gannet Hallar, A., Stratmann, F., and Bilde, M.: Hygroscopic growth and CCN  
686 activity of HULIS from different environments, *J. Geophys. Res. Atmos.*, 117, 22203, <https://doi.org/10.1029/2012JD018249>,  
687 2012.

688 Liu, J. and Li, Z.: Significant Underestimation in the Optically Based Estimation of the Aerosol First Indirect Effect Induced  
689 by the Aerosol Swelling Effect, *Geophys. Res. Lett.*, 45, 5690–5699, <https://doi.org/10.1029/2018GL077679>, 2018.

690 Liu, X. and Wang, J.: How important is organic aerosol hygroscopicity to aerosol indirect forcing?,  
691 <https://doi.org/10.1088/1748-9326/5/4/044010>, 2010.

692 Magee Scientific: User’s manual for Aethalometer® Model AE33, 149, 2018.

693 Maskey, S., Chong, K. Y., Kim, G., Kim, J. S., Ali, A., and Park, K.: Effect of mixing structure on the hygroscopic behavior  
694 of ultrafine ammonium sulfate particles mixed with succinic acid and levoglucosan, *Particuology*, 13, 27–34,  
695 <https://doi.org/10.1016/J.PARTIC.2013.08.004>, 2014.

696 Massoli, P., Bates, T. S., Quinn, P. K., Lack, D. A., Baynard, T., Lerner, B. M., Tucker, S. C., Brioude, J., Stohl, A., and  
697 Williams, E. J.: Aerosol optical and hygroscopic properties during TexAQS-GoMACCS 2006 and their impact on aerosol  
698 direct radiative forcing, *J. Geophys. Res. Atmos.*, 114, 0–07, <https://doi.org/10.1029/2008JD011604>, 2009.

699 Mbengue, S., Zikova, N., Schwarz, J., Vodička, P., Šmejkalová, A. H., and Holoubek, I.: Mass absorption cross-section and  
700 absorption enhancement from long term black and elemental carbon measurements: A rural background station in Central  
701 Europe, *Sci. Total Environ.*, 794, 1–14, <https://doi.org/10.1016/j.scitotenv.2021.148365>, 2021.

702 Müller, T. and Fiebig, M.: ACTRIS In Situ Aerosol: Guidelines for Manual QC of AE33 absorption photometer data, 9 pp.,  
703 2021.

704 Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza,  
705 B., Nakajima, T., Robock, A., Stephens, T., Takemura, T., and Zhang, H.: Anthropogenic and natural radiative forcing, in:  
706 *Climate Change 2013 the Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the*  
707 *Intergovernmental Panel on Climate Change*, vol. 9781107057, edited by: Stocker, T. F., Qin, D. G.-K. P., Tignor, M., Allen,  
708 S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, United  
709 Kingdom and New York, NY, USA, 659–740, <https://doi.org/10.1017/CBO9781107415324.018>, 2013.

710 HYSPLIT model: <https://www.ready.noaa.gov/HYSPLIT.php>.

711 Orozco, D., Beyersdorf, A. J., Ziemba, L. D., Berkoff, T., Zhang, Q., Delgado, R., Hennigan, C. J., Thornhill, K. L., Young,  
712 D. E., Parworth, C., Kim, H., and Hoff, R. M.: Hygroscopicity measurements of aerosol particles in the San Joaquin Valley,  
713 CA, Baltimore, MD, and Golden, CO, *J. Geophys. Res. Atmos.*, 121, 7344–7359, <https://doi.org/10.1002/2015JD023971>,  
714 2016.

715 Pandolfi, M., Alados-Arboledas, L., Alastuey, A., Andrade, M., Angelov, C., Artiñano, B., Backman, J., Baltensperger, U.,  
716 Bonasoni, P., Bukowiecki, N., Collaud Coen, M., Conil, S., Coz, E., Crenn, V., Dudoitis, V., Ealo, M., Eleftheriadis, K., Favez,  
717 O., Fetfatzis, P., Fiebig, M., Flentje, H., Ginot, P., Gysel, M., Henzing, B., Hoffer, A., Holubova Smejkalova, A., Kalapov, I.,  
718 Kalivitis, N., Kouvarakis, G., Kristensson, A., Kulmala, M., Lihavainen, H., Lunder, C., Luoma, K., Lyamani, H., Marinoni,  
719 A., Mihalopoulos, N., Moerman, M., Nicolas, J., O’Dowd, C., Petäjä, T., Petit, J. E., Marc Pichon, J., Prokopciuk, N., Putaud,  
720 J. P., Rodríguez, S., Sciare, J., Sellegri, K., Swietlicki, E., Titos, G., Tuch, T., Tunved, P., Ulevicius, V., Vaishya, A., Vana,  
721 M., Virkkula, A., Vratolis, S., Weingartner, E., Wiedensohler, A., and Laj, P.: A European aerosol phenomenology - 6:  
722 Scattering properties of atmospheric aerosol particles from 28 ACTRIS sites, *Atmos. Chem. Phys.*, 18, 7877–7911,  
723 <https://doi.org/10.5194/acp-18-7877-2018>, 2018.

724 Perrone, M. R., Romano, S., Genga, A., and Paladini, F.: Integration of optical and chemical parameters to improve the  
725 particulate matter characterization, *Atmos. Res.*, 205, 93–106, <https://doi.org/10.1016/j.atmosres.2018.02.015>, 2018.

726 Pilat, M. J. and Charlson, R. J.: Theoretical and optical studies of humidity effects on the size distribution of a hygroscopic  
727 aerosol, *J. Rech. atmosphériques*, 165–170, 1966.

728 Pöhlker, M. L., Pöhlker, C., Quaas, J., Mülmenstädt, J., Pozzer, A., Andreae, M. O., Artaxo, P., Block, K., Coe, H., Ervens,  
729 B., Gallimore, P., Gaston, C. J., Gunthe, S. S., Henning, S., Herrmann, H., Krüger, O. O., McFiggans, G., Poulain, L., Raj, S.  
730 S., Reyes-Villegas, E., Royer, H. M., Walter, D., Wang, Y., and Pöschl, U.: Global organic and inorganic aerosol

731 hygroscopicity and its effect on radiative forcing, *Nat. Commun.* 2023 141, 14, 6139-, [https://doi.org/10.1038/s41467-023-](https://doi.org/10.1038/s41467-023-41695-8)  
732 41695-8, 2023.

733 Ray, A., Bhowmik, M., Hazra, A., and Pandithurai, G.: The influence of aerosol hygroscopicity on clouds and precipitation  
734 over Western Ghats, India, *Q. J. R. Meteorol. Soc.*, 150, 1473–1488, <https://doi.org/10.1002/QJ.4654>, 2024.

735 Ren, R., Li, Z., Yan, P., Wang, Y., Wu, H., Cribb, M., Wang, W., Jin, X., Li, Y., and Zhang, D.: Measurement report: The  
736 effect of aerosol chemical composition on light scattering due to the hygroscopic swelling effect, *Atmos. Chem. Phys.*, 21,  
737 9977–9994, <https://doi.org/10.5194/ACP-21-9977-2021>, 2021.

738 Savadkoohi, M., Pandolfi, M., Favez, O., Putaud, J. P., Eleftheriadis, K., Fiebig, M., Hopke, P. K., Laj, P., Wiedensohler, A.,  
739 Alados-Arboledas, L., Bastian, S., Chazneau, B., María, Á. C., Colombi, C., Costabile, F., Green, D. C., Hueglin, C., Liakakou,  
740 E., Luoma, K., Listrani, S., Mihalopoulos, N., Marchand, N., Močnik, G., Niemi, J. V., Ondráček, J., Petit, J. E., Rattigan, O.  
741 V., Reche, C., Timonen, H., Titos, G., Tremper, A. H., Vratolis, S., Vodička, P., Funes, E. Y., Zíková, N., Harrison, R. M.,  
742 Petäjä, T., Alastuey, A., and Querol, X.: Recommendations for reporting equivalent black carbon (eBC) mass concentrations  
743 based on long-term pan-European in-situ observations, *Environ. Int.*, 185, 108553,  
744 <https://doi.org/10.1016/J.ENVINT.2024.108553>, 2024.

745 Savadkoohi, M., Gherras, M., Favez, O., Petit, J. E., Rovira, J., Chen, G. I., Via, M., Platt, S., Aurela, M., Chazneau, B., de  
746 Brito, J. F., Riffault, V., Eleftheriadis, K., Flentje, H., Gysel-Beer, M., Hueglin, C., Rigler, M., Gregorič, A., Ivančič, M.,  
747 Keernik, H., Maasikmets, M., Liakakou, E., Stavroulas, I., Luoma, K., Marchand, N., Mihalopoulos, N., Petäjä, T., Prevot, A.  
748 S. H., Daellenbach, K. R., Vodička, P., Timonen, H., Tobler, A., Vasilescu, J., Dandocsi, A., Mbengue, S., Vratolis, S.,  
749 Zografou, O., Chauvigné, A., Hopke, P. K., Querol, X., Alastuey, A., and Pandolfi, M.: Addressing the advantages and  
750 limitations of using Aethalometer data to determine the optimal absorption Ångström exponents (AAEs) values for eBC source  
751 apportionment, *Atmos. Environ.*, 349, <https://doi.org/10.1016/j.atmosenv.2025.121121>, 2025.

752 Seinfeld, J. H. and Pandis, S. N.: *Atmospheric chemistry and physics*, Second Edi., John Wiley & Sons, Ltd, New Jersey, 1203  
753 pp., [https://doi.org/10.1007/978-3-662-04540-4\\_16](https://doi.org/10.1007/978-3-662-04540-4_16), 2006.

754 Shen, Y., Zhi, G., Zhang, Y., Jin, W., Kong, Y., Li, Z., and Zhang, H.: An investigative review of the expanded capabilities of  
755 thermal/optical techniques for measuring carbonaceous aerosols and beyond, *Environ. Pollut.*, 364, 125363,  
756 <https://doi.org/10.1016/J.ENVPOL.2024.125363>, 2025.

757 Song, M., Liu, P. F., Hanna, S. J., Zaveri, R. A., Potter, K., You, Y., Martin, S. T., and Bertram, A. K.: Relative humidity-  
758 dependent viscosity of secondary organic material from toluene photo-oxidation and possible implications for organic  
759 particulate matter over megacities, *Atmos. Chem. Phys.*, 16, 8817–8830, <https://doi.org/10.5194/ACP-16-8817-2016>, 2016.

760 Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's HYSPLIT atmospheric  
761 transport and dispersion modeling system, *Bull. Am. Meteorol. Soc.*, 96, 2059–2077, [https://doi.org/10.1175/BAMS-D-14-](https://doi.org/10.1175/BAMS-D-14-00110.1)  
762 00110.1, 2015.

763 Suchánková, L., Crumeyrolle, S., Bourrienne, E., Prokeš, R., Holoubek, I., Ždímal, V., and Chiapello, I.: An insight into recent  
764 PM1 aerosol light scattering properties and particle number concentration variabilities at the suburban site ATOLL in Northern  
765 France, *Sci. Total Environ.*, 959, 178190, <https://doi.org/10.1016/J.SCITOTENV.2024.178190>, 2025.

766 Tang, J., Li, J., Mo, Y., Safaei Khorrām, M., Chen, Y., Tang, J., Zhang, Y., Song, J., and Zhang, G.: Light absorption and  
767 emissions inventory of humic-like substances from simulated rainforest biomass burning in Southeast Asia, *Environ. Pollut.*,  
768 262, <https://doi.org/10.1016/J.ENVPOL.2020.114266>, 2020.

769 Thomsen, D., Iversen, E. M., Skønager, J. T., Luo, Y., Li, L., Roldin, P., Priestley, M., Pedersen, H. B., Hallquist, M., Ehn,  
770 M., Bilde, M., and Glasius, M.: The effect of temperature and relative humidity on secondary organic aerosol formation from  
771 ozonolysis of  $\Delta^3$ -carene, *Environ. Sci. Atmos.*, 4, 88–103, <https://doi.org/10.1039/D3EA00128H>, 2024.

772 Titos, G., Lyamani, H., Cazorla, A., Sorribas, M., Foyo-Moreno, I., Wiedensohler, A., and Alados-Arboledas, L.: Study of the  
773 relative humidity dependence of aerosol light-scattering in southern Spain, *Tellus B Chem. Phys. Meteorol.*, 66, 24536,  
774 <https://doi.org/10.3402/tellusb.v66.24536>, 2014.

775 Titos, G., Cazorla, A., Zieger, P., Andrews, E., Lyamani, H., Granados-Muñoz, M. J., Olmo, F. J., and Alados-Arboledas, L.:  
776 Effect of hygroscopic growth on the aerosol light-scattering coefficient: A review of measurements, techniques and error  
777 sources, *Atmos. Environ.*, 141, 494–507, <https://doi.org/10.1016/j.atmosenv.2016.07.021>, 2016.

778 Titos, G., Burgos, M. A., Zieger, P., Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Weingartner, E.,  
779 Henzing, B., Luoma, K., O'Dowd, C., Wiedensohler, A., and Andrews, E.: A global study of hygroscopicity-driven light-  
780 scattering enhancement in the context of other in situ aerosol optical properties, *Atmos. Chem. Phys.*, 21, 13031–13050,  
781 <https://doi.org/10.5194/acp-21-13031-2021>, 2021.

782 Toll, V., Christensen, M., Quaas, J., and Bellouin, N.: Weak average liquid-cloud-water response to anthropogenic aerosols,  
783 *Nat.* 2019 5727767, 572, 51–55, <https://doi.org/10.1038/s41586-019-1423-9>, 2019.

784 Vodička, P., Schwarz, J., and Ždímal, V.: Analysis of one year's OC/EC data at a Prague suburban site with 2-h time resolution,  
785 *Atmos. Environ.*, 77, 865–872, <https://doi.org/10.1016/j.atmosenv.2013.06.013>, 2013.

786 Vodička, P., Schwarz, J., Cusack, M., and Ždímal, V.: Detailed comparison of OC/EC aerosol at an urban and a rural Czech  
787 background site during summer and winter, *Sci. Total Environ.*, 518–519, 424–433,  
788 <https://doi.org/10.1016/j.scitotenv.2015.03.029>, 2015.

789 Watson-Parris, D. and Smith, C. J.: Large uncertainty in future warming due to aerosol forcing, *Nat. Clim. Chang.* 2022 1212,  
790 12, 1111–1113, <https://doi.org/10.1038/s41558-022-01516-0>, 2022.

791 Williams, A. I. L., Stier, P., Dagan, G., and Watson-Parris, D.: Strong control of effective radiative forcing by the spatial  
792 pattern of absorbing aerosol, *Nat. Clim. Chang.* 2022 128, 12, 735–742, <https://doi.org/10.1038/s41558-022-01415-4>, 2022.

793 Wiscombe, W. J. and Grams, G. W.: Backscattered fraction in two-stream approximations, *J. Atmos. Sci.*, 33, 2440–2451,  
794 [https://doi.org/10.1175/1520-0469\(1976\)033<2440:tbfits>2.0.co;2](https://doi.org/10.1175/1520-0469(1976)033<2440:tbfits>2.0.co;2), 1976.

795 WMO/GAW: WMO/GAW Aerosol Measurement Procedures, Guidelines and Recommendations (2nd Edition) | Climate &  
796 Clean Air Coalition, Geneva, 103 pp., 2016.

797 Wu, T. and Boor, B. E.: Urban aerosol size distributions: A global perspective, *Atmos. Chem. Phys.*, 21, 8883–8914,  
798 <https://doi.org/10.5194/acp-21-8883-2021>, 2021.

799 Zelinka, M. D., Smith, C. J., Qin, Y., and Taylor, K. E.: Comparison of methods to estimate aerosol effective radiative forcings  
800 in climate models, *Atmos. Chem. Phys.*, 23, 8879–8898, <https://doi.org/10.5194/ACP-23-8879-2023>, 2023.

801 Zhang, J., Chen, Y. S., Gryspeerdt, E., Yamaguchi, T., and Feingold, G.: Radiative forcing from the 2020 shipping fuel  
802 regulation is large but hard to detect, *Commun. Earth Environ.* 2025 61, 6, 1–11, [https://doi.org/10.1038/s43247-024-01911-](https://doi.org/10.1038/s43247-024-01911-9)  
803 9, 2025.

804 Zieger, P., Fierz-Schmidhauser, R., Poulain, L., Müller, T., Birmili, W., Spindler, G., Wiedensohler, A., Baltensperger, U.,  
805 and Weingartner, E.: Influence of water uptake on the aerosol particle light scattering coefficients of the Central European

806 aerosol, *Tellus B Chem. Phys. Meteorol.*, 66, <https://doi.org/10.3402/TELLUSB.V66.22716>, 2014.

807 Zieger, P., Aalto, P. P., Aaltonen, V., Äijälä, M., Backman, J., Hong, J., Komppula, M., Krejci, R., Laborde, M., Lampilahti,  
808 J., De Leeuw, G., Pfüller, A., Rosati, B., Tesche, M., Tunved, P., Väänänen, R., and Petäjä, T.: Low hygroscopic scattering  
809 enhancement of boreal aerosol and the implications for a columnar optical closure study, *Atmos. Chem. Phys.*, 15, 7247–  
810 7267, <https://doi.org/10.5194/acp-15-7247-2015>, 2015.

811