



1 **A European aerosol phenomenology-6: Scattering properties of atmospheric aerosol**
2 **particles from 28 ACTRIS sites**

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1 **Abstract**

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4 This paper presents the light scattering properties of atmospheric aerosol particles measured over
5 the past decade at 28 ACTRIS observatories, located mainly in Europe. The data include particle
6 light scattering (σ_{sp}) and hemispheric backscattering (σ_{bsp}) coefficients, scattering Ångström
7 exponent (SAE), backscatter fraction (BF) and asymmetry parameter (g). A large range of σ_{sp} was
8 observed across the network. Low σ_{sp} values were on average measured in Nordic and Baltic
9 countries and in Western Europe whereas the highest σ_{sp} were measured at regional sites in
10 eastern and central Europe. In these regional areas the SAE was also high indicating the
11 predominance of fine-mode particles. On average, the SAE was lower in the Nordic and Baltic,
12 western and southern countries suggesting a lower fraction of fine-mode particle compared to
13 central and eastern Europe. An increasing gradient of σ_{sp} was observed when moving from
14 mountain to regional and to urban sites. Conversely, the mass-independent SAE and g parameters
15 did not show the same gradient. At all sites, both SAE and g varied greatly with aerosol particle
16 loading. The lowest values of g were always observed under low σ_{sp} indicating a larger contribution
17 from particles in the smaller accumulation mode. Then, g steeply increased with increasing σ_{sp}
18 indicating a progressive shift of the particle size distribution toward the larger end of the
19 accumulation mode. Under periods of high particle mass concentrations, the variation of g was less
20 pronounced whereas the SAE increased or decreased suggesting changes mostly in the coarse
21 aerosol particles mode rather than in the fine mode. The station placement seemed to be the main
22 parameter affecting the intra-annual variability. At mountain sites, higher σ_{sp} was measured in
23 summer mainly because of the enhanced boundary layer influence. Conversely, less horizontal
24 and vertical dispersion in winter led to higher σ_{sp} at all low altitude sites in central and eastern
25 Europe compared to summer. On average, these sites also showed SAE maxima in summer (and
26 correspondingly g minima). Large intra-annual variability of SAE and g was observed also at
27 Nordic and Baltic countries due to seasonal-dependent transport of different air masses to these
28 remote sites. Statistically significant decreasing trends of σ_{sp} were observed at 5 out of 13 stations
29 included in trend analyses. The total reductions of σ_{sp} were consistent with those reported for $PM_{2.5}$
30 and PM_{10} mass concentrations over similar periods across Europe.

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1 1. Introduction

2 Atmospheric aerosol particles are recognized as an important atmospheric constituent with
3 demonstrated effects on climate and health. Radiative forcing of aerosol particles, estimated as
4 -0.9 [-1.9 to -0.1] W/m^2 (IPCC, 2014), has two competing components: a cooling effect from most
5 particle types and a partially offsetting warming contribution from black carbon (BC) particle light
6 absorption of solar radiation. The aerosol cooling is the dominant effect; thus aerosol particles are
7 counteracting a substantial portion of warming effect from well mixed greenhouse gases (GHGs).
8 This process is driven by the scattering properties of most aerosol particle types (e.g. secondary
9 sulfate and nitrate particles, mineral and organic matter), which reduces the amount of solar
10 radiation reaching the Earth surface reflecting it back to space and modifying the Earth's radiative
11 balance. However, the high temporal and spatial variability of atmospheric aerosol particles, due to
12 the wide variety of aerosol sources and sinks, their short and variable lifetime (hours to weeks in
13 the planetary boundary layer) and spatial non-uniformity, contribute to the largest uncertainty in the
14 estimation of the total radiative forcing. Reducing these uncertainties is mandatory in view of global
15 warming experienced over the past 50 years. In fact, there are evidences suggesting that the
16 observed (and projected) decrease in emissions of anthropogenic aerosol particles, in response to
17 air quality policies, would eventually unmask the global warming (Rotstajn et al., 2013). Thus,
18 current emission controls could increase climate warming while improving air quality (e.g. Stohl
19 et al., 2015). The measurements of aerosol particle optical properties such as light scattering and
20 absorption, together with measurements of physical and chemical properties, are fundamental in
21 order to better understand the current conflict involving a trade-off between the impacts of aerosols
22 on environmental health and Earth's climate. Several international projects are providing in the last
23 decades important information on the atmospheric particle properties worldwide. Near-surface in-
24 situ observations of aerosol particle properties are performed worldwide under the GAW/WMO
25 program completed with policy-oriented programs such as IMPROVE (Interagency Monitoring of
26 Protected Visual Environments; <http://vista.cira.colostate.edu/Improve/>) in USA or EMEP
27 (European Monitoring and Evaluation Programme; <http://www.emep.int/>) in Europe. Additional
28 information specifically targeting advanced aerosol particle properties are obtained in Europe using
29 information from the European research infrastructure ACTRIS (Aerosols, Clouds, and Trace
30 gases Research InfraStructure; <http://www.actris.eu>) or from short-term RTD projects such as
31 EUCAARI (European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions;
32 <http://www.cas.manchester.ac.uk/resprojects/eucaari/>). The implementation of the GAW program
33 in Europe is performed under ACTRIS for the advanced observation of aerosol particle properties.
34 ACTRIS is providing harmonized measurement of different (physical, chemical and optical) aerosol
35 properties in a systematic way at major observation sites in Europe. More than 60 measuring sites
36 worldwide are currently providing ground-based in-situ aerosol particle light scattering
37 measurements (EBAS database; [www. http://ebas.nilu.no/](http://ebas.nilu.no/)) and the number has increased
38 substantially in the last decade.



1 The objective of this work is to integrate the total aerosol light scattering coefficient (σ_{sp}) and
2 hemispheric backscattering coefficient (σ_{bsp}) measurements performed over several years at the
3 ground based in-situ ACTRIS stations. A total of 28 stations (25 European + 3 non-European) are
4 included in order to document the variability in near-surface aerosol particle light scattering across
5 the ACTRIS network. Moreover, at some of the ACTRIS stations more than 10 years of σ_{sp} data
6 are available allowing us to perform trends analysis. The study of the trend of σ_{sp} is important given
7 that decreasing or increasing trend of σ_{sp} would mirror the effectiveness of the air quality control
8 measures. In fact, many studies have shown that the concentrations of particulate matter (PM),
9 and other air pollutants such as sulfur dioxide (SO₂) and carbon monoxide (CO), have clearly
10 decreased during the last 20 years in many European Countries (Barmpadimos et al., 2012;
11 Cusack et al., 2012; EEA, 2013; Querol et al., 2014; Guerreiro et al., 2014; Pandolfi et al., 2016,
12 Tørseth et al., 2012, among others).

13 Previous studies presenting multi-site ground-based in-situ aerosol particle optical measurements
14 were for example performed by Delene and Ogren (2002), Collaud Coen et al. (2013) and Andrews
15 et al. (2011). Delene and Ogren (2002) reported the variability of aerosol particle optical properties
16 at four North American surface monitoring sites. Collaud Coen et al. (2013) presented long term
17 (>8-9 years) aerosol particle light scattering and absorption measurements performed at 24
18 regional/remote observatories, among which 5 of them are located in Europe. Andrews et al.
19 (2011) reported the aerosol particle optical measurements performed at 12 (4 located in Europe)
20 mountain top observatories. Thus, the number of papers reporting aerosol particle optical
21 properties measured at different sites is rather scarce and unfortunately almost inexistent outside
22 Europe and the United States.

23 Our work is focused mainly on European observatories aiming at a representative phenomenology
24 of aerosol particle light scattering coefficient measured at ACTRIS stations. Thanks to the
25 establishment of European monitoring networks and/or research projects five papers have been
26 published related with aerosol phenomenology in Europe: Van Dingenen et al. (2004) and Putaud
27 et al. (2004) on the physical and chemical, respectively, characteristics of particulate matter (PM)
28 at kerbside, urban, rural and background sites in Europe; Putaud et al. (2010) on the physical and
29 chemical characteristics of PM measured at 60 sites across Europe; Cavalli et al. (2016) on the
30 harmonized concentrations of carbonaceous aerosol at ten regional background sites in Europe;
31 Zanatta et al. (2016) presenting a climatology of BC optical properties at nine European regional
32 background sites. The importance of these studies and of the present work relies on the evidence
33 that a reliable assessment of the physical, chemical and optical properties of aerosol particles at
34 European scale is of crucial importance for an accurate estimation of the radiative forcing of
35 atmospheric aerosols. This work is the first European phenomenology study dedicated to the light
36 scattering properties of aerosol particles measured in-situ at near-surface ground-based
37 observatories. Moreover, the trend analyses presented can be used to evaluate how the European
38 mitigation strategies adopted to improve air quality affected the aerosol particle optical properties.



1 In fact, starting from the σ_{sp} measurements performed at the ACTRIS observatories, three
2 intensive aerosol particle optical parameters can be estimated, namely the scattering Ångström
3 exponent (SAE), the backscattering fraction (BF), and the asymmetry parameter (g). These
4 intensive properties do not depend on the PM mass concentration and are directly related to
5 aerosol particles properties such as size, shape, size distribution and chemical composition. The
6 SAE can be considered a proxy for the aerosol particle size range with higher (lower) SAE
7 associated to predominance of fine (course) aerosol particles (e.g. Seinfeld and Pandis, 1998;
8 Esteve et al., 2012; A. Valenzuela et al., 2015 among others). The BF and g parameters are
9 calculated quantities that influence the variability of the radiative forcing efficiency and that
10 represent the angular light scattering of aerosol particles. For computational efficiency, the angular
11 light scattering is often represented by a single value (BF, σ_{sp}/σ_{bsp} or g) (Ogren et al., 2006). For
12 some of the ACTRIS data used in this work, trends of these intensive aerosol particle optical
13 properties are investigated as well.

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15 **2. Experimental**

16 **2.1 Atmospheric Observatories**

17 Figure 1 shows the location of the observatories which are grouped based on their geographical
18 location as performed in other European phenomenology studies (e.g. Putaud et al., 2010).
19 Observatories information and measurements periods are summarized in Table 1. The
20 observatories are also divided in five different categories depending on their placement in each
21 geographical sector. Mountain: includes those observatories located at more than 1 km above sea
22 level; coastal: includes observatories located close to the sea coast; regional: includes those
23 observatories mostly affected by regional sources and closer to large pollution sources compared
24 to continental sites; continental: comprise observatories located in remote continental areas;
25 urban/sub-urban: includes observatories located in urban background or suburban areas.

26 Nordic and Baltic stations are represented by Birkenes (BIR, Norway; regional), Hyytiälä (SMR,
27 Finland; regional), Pallas (PAL, Finland; continental), Vavihill (VHL, Sweden; continental), and
28 Preila (PLA, Lithuania; coastal). Western European sites are Puy De Dome (PUY, France;
29 mountain), Mace Head (MHD, Ireland; coastal), Cabauw (CBW, The Netherlands; regional), SIRTA
30 (SIR, France; suburban), and Observatory Perenne (OPE, France; regional). Central European
31 stations are Jungfraujoch (JFJ, Switzerland; mountain), Hohenpeissenberg (HPB, Germany;
32 mountain), Melpitz (MPZ, Germany; regional), Ispra (IPR, Italy; semi regional), Mt. Cimone (CMN,
33 Italy; mountain) and Košetice (KOS, Czech Republic; regional). Eastern European stations are Beo
34 Moussala (BEO, Bulgaria; mountain) and K-Pusztá (KPS, Hungary; regional). South-western
35 European stations are represented by Izaña (IZO, Spain; mountain), Montsec (MSA, Spain;
36 mountain), Montseny (MSY, Spain; regional), Madrid (MAD, Spain; sub-urban), and Granada



1 (UGR, Spain; urban) whereas south-eastern European stations are Athens (DEM, Greece; sub-
2 urban) and Finokalia (FKL, Greece; coastal). Finally, Arctic and Antarctic stations are Zeppelin
3 (ZEP) and Troll (TRL), respectively. Another non-European mountain station included is Mt.
4 Chacaltaya (CHC, Bolivia; mountain). The altitude of the mountain stations considered here ranges
5 between 985 m at HPB to 5240 m at CHC. Some of the mountain stations included in this
6 investigation have been already included in the work by Andrews et al. (2011), namely IZO, JFJ,
7 CMN, and BEO. Moreover, FKL, HPB, JFJ, MHD, and PAL stations have been included in the
8 study by Collaud Coen et al. (2013). Both studies presented in-situ aerosol particle optical
9 measurements performed at these stations. Main results from the previous investigation are
10 summarized in the results section.

11 At JFJ, HPB, IPR (Central Europe), UGR (southwestern Europe), MHD (western Europe), PAL and
12 SMR (Nordic and Baltic), at least 10 years of data are available for trend analysis. However, in
13 order to improve the spatial coverage, trends are also studied at CMN, MPZ (central Europe), IZO
14 (southwestern Europe), PUY (western Europe), KPS and BEO (eastern Europe), where >8-9 years
15 of data are available (cf. Table 1). The stations included in this work report the data to ACTRIS and
16 GAW/EMEP, consequently the data are quality assured given that the nephelometer instruments
17 are run following the ACTRIS/GAW standards (WMO-GAW Report, 2016) and regularly inter-
18 compared.

19

20 **2.2 Scattering measurements**

21 **2.2.1 Instruments**

22 The measurements of σ_{sp} and σ_{bsp} included in this study were obtained from TSI and Ecotech
23 integrating nephelometers (Table 1). These optical instruments measure the amount of light
24 scattered by particles in the visible spectrum and provide σ_{sp} and σ_{bsp} coefficients of sampled
25 aerosols. Most used nephelometer models are the TSI3563 and the Ecotech AURORA3000, both
26 providing σ_{sp} and σ_{bsp} . Model TSI3563 measures σ_{sp} and σ_{bsp} at 450, 550, and 700 nm whereas the
27 Ecotech AURORA3000 measures at 450, 525 and 635 nm. Other used models are the M9003
28 from Ecotech (SIR and CMN) and the RR (Radiance Research) nephelometer model M903 (FKL)
29 measuring σ_{sp} at 520 nm and 532 nm, respectively. Due to the non-homogeneity of the light source
30 of the model M9003, the light source was changed at SIR in 2013 with the AURORA3000 light
31 source and at CMN in 2009 with an opal glass light source. The detailed description of the main
32 characteristics and working principle of the integrating nephelometers can be found e.g. in Müller
33 et al. (2011) for the Ecotech AURORA3000 and in Anderson and Ogren (1998) for the model TSI
34 3563. Following the ACTRIS and WMO-GAW recommendations, the nephelometers are regularly
35 calibrated using span gas and zero-adjusted using particle-free air. Recommended quality
36 assurance procedures during on-site operation as described in GAW (WMO/GAW, 2016),
37 guarantee the quality and comparability of the data. Moreover, most of the integrating



1 nephelometers involved in ACTRIS have undergone performance checks at scheduled times at the
2 World Calibration Center for Aerosol Physical properties of ACTRIS/GAW.

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4 **2.2.2 Data treatment**

5 **2.2.2.1 Truncation correction**

6 Data from integrating nephelometers used here are corrected for non-ideal illumination of the light
7 source (deviation from Lambertian distribution of light) and for truncation of the sensing volumes in
8 the near-forward (around 0-10°) and near-backward direction (around 170-180°) (Müller et al.,
9 2009 and Anderson and Ogren, 1998). Correction schemes have been provided by Müller et al.
10 (2011) for the Ecotech AURORA3000 and by Anderson and Ogren (1998) for the TSI3563. Both
11 methods provide a simple linear correction scheme based on the scattering Ångström exponent
12 (SAE) determined from raw nephelometer data to correct for the size distribution-dependent
13 truncation error. It has been demonstrated that for an aerosol particle population with a single
14 scattering albedos (SSA) greater than 0.8 this simple correction scheme provides a suitable
15 quantification of the truncation error (Müller et al., 2011). However, for SSA < 0.8 a correction
16 scheme based on particle number size distribution should be used (Müller et al., 2011). The
17 aerosol particle light scattering data used here are corrected for non-ideal illumination and for
18 truncation by the data providers or in this work. This information is reported in Table S1 of the
19 Supporting Material. Only at SIR, FKL, and CMN, σ_{sp} data are not corrected for truncation because
20 σ_{sp} at these observatories was measured at one wavelength. At CMN, the 3- λ TSI3563 is operative
21 since 2014 (cf. Table 1). However, not correcting for truncation doesn't prevent from comparing
22 non-corrected σ_{sp} with truncation corrected σ_{sp} . The truncation correction increases with particle
23 size being more important for coarse aerosol particles. For example, using the SAE calculated at
24 CMN for the years 2014-2015 and the correction scheme provided for the TSI3563 by Anderson
25 and Ogren (1998), the difference between non-corrected σ_{sp} and corrected mean σ_{sp} is lower than
26 7% at this site.

27

28 **2.2.2.2 Relative humidity**

29 The integrating nephelometer measurements within ACTRIS and WMO-GAW should be performed
30 at low relative humidity (RH) in order to avoid enhanced scattering due to water uptake of aerosol
31 particles and to make measurements comparable. For the Ecotech integrating nephelometers the
32 RH threshold can be set by using a processor-controlled automatic heater inside the
33 nephelometer. At some mountain sites where whole air is sampled (cf. Table 1), the natural
34 temperature difference between outside and inside air dries cloud droplets to the aerosol phase
35 when a cloud is present at the station. RH is also controlled by de-humidifying in the inlet pipe as



1 reported in the GAW report 226 to ensure sampling RH of less than 40%. This recommendation
2 intends to make the data comparable across the network, which otherwise would be a strong
3 function of the highly variable sample RH. Currently, at the majority of ACTRIS observatories, the
4 aerosol particles light scattering measurements are performed at RH lower than 40%. However,
5 given that at some stations the 40% RH threshold is sometimes exceeded, we selected in this work
6 a RH threshold of 50% in order to improve the data coverage. Estimating the aerosol particle light
7 scattering enhancement due to an increase of RH from 40% to 50% is difficult using the data
8 available here because σ_{sp} measurements at RH>40% are not evenly distributed over the
9 measurement periods. In fact, at the majority of the stations RH higher than 40% is registered
10 mostly in summer. However, the scattering enhancement due to a change in RH between 40% and
11 50% should be small and will not exceed around 3-5% even for more hygroscopic particles (e.g.
12 Fierz-Schmidhauser et al., 2010a,b). Table S2 in the Supporting Material reports the number of RH
13 hourly data reported at each observatory and the number and % of hourly RH data >50%. The
14 frequency distributions of measured RH are shown in Figure S1. Finally, σ_{sp} and σ_{bsp} data reported
15 to EBAS and used in this work are referenced to standard T (273.15 °C) and P (1013 hPa)
16 conditions.

17 **2.2.2.3 Available wavelengths**

18 In this work we present and discuss the σ_{sp} , BF and g measurements obtained using the green
19 wavelength of the integrating nephelometers. The available wavelengths ranged from 520 nm (2
20 stations, CMN and VHL) to 550 nm (18 stations). Other used wavelengths are 525 nm (6 stations),
21 532 nm (used at FKL until 2010; cf. Table 2). An exception is SIR where only σ_{sp} at 450 nm is
22 available. The measurements of σ_{sp} reported here are not adjusted to 550 nm which generally is
23 the most used wavelength (e.g. Andrews et al., 2011) because of the different data availability of
24 σ_{sp} and SAE at the measuring stations. As discussed in the following Sections SAE is calculated
25 for σ_{sp} data higher than 0.8 Mm^{-1} , thus leading to different data coverage for σ_{sp} and SAE and thus
26 preventing the adjustment of all measured σ_{sp} to 550 nm. Moreover, SAE is not available at FKL
27 and SIR (and at CMN until 2014) thus preventing any wavelength adjustment at these stations.
28 Using the mean SAE calculated at those stations where σ_{sp} is measured at different wavelength
29 than 550 nm (cf. Tables S4 and S5 in Supporting material), we estimate differences in σ_{sp} lower
30 than 6% after adjusting to 550 nm. At FKL and SIR, where SAE is not available and assuming a
31 SAE of 1.5, the difference by adjusting to 550 nm is 4.9% at FKL and 26% at SIR, respectively.
32 The higher difference at SIR is due to the fact that measurements at this station are performed at
33 450 nm. Finally, at CMN the effect of the adjustment of σ_{sp} to 550 nm (from 520 nm) using a mean
34 SAE of 2 (cf. Table S5) is lower than 10%.

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1 2.2.3 Calculation of aerosol particle intensive optical properties

2 In addition to the direct σ_{sp} and σ_{bsp} measurements obtained with the above detailed
3 instrumentation, the following aerosol intensive parameters are calculated from hourly-averaged in-
4 situ data.

5 The scattering Ångström exponent (SAE) characterizes the wavelength dependency of σ_{sp} and it
6 can be calculated as follows (with $\lambda_1 > \lambda_2$):

$$7 \quad SAE = - \frac{\log\left(\frac{\sigma_{sp}^{\lambda_1}}{\sigma_{sp}^{\lambda_2}}\right)}{\log\left(\frac{\lambda_1}{\lambda_2}\right)} \quad (\text{Eq. 1})$$

8 Here, the SAE is calculated as linear estimation of σ_{sp} measured at the three available
9 wavelengths. The SAE depends on particle size distribution. It takes values greater than 2 when
10 the light scattering is dominated by fine particles (radii $\leq 0.5 \mu\text{m}$ as e.g. in Schuster et al. (2006)),
11 while it is lower than one when the light scattering is increasingly dominated by coarse particles
12 (Seinfeld and Pandis, 1998; Schuster et al., 2006).

13 The asymmetry parameter (g) (Andrews et al., 2006; Delene and Ogren, 2002) describes the
14 probability that the radiation is scattered in a given radiation and it is defined as the cosine-
15 weighted average of the phase function. Thus, g gives information on the amount of radiation that
16 a particle can scatter in the forward direction compared to the backward direction. Theoretically,
17 the values of g can range from -1 for only back scattering to $+1$ for complete forward scattering
18 (0°), with a value of 0.7 commonly used in radiative transfer models (Ogren et al., 2006). The g
19 parameter can be estimated from the backscatter fraction (BF) which is the ratio between σ_{bsp} and
20 σ_{sp} (Andrews et al., 2006):

$$21 \quad g = -7.14(BF)^3 + 7.46(BF)^2 - 3.96(BF) + 0.9893 \quad (\text{Eq. 2})$$

22

23 2.2.4 Data coverage

24 Table S3 in the Supporting Material reports the number of hours and data availability for each
25 atmospheric observatory. The data coverage reported in Table S3, refers to scattering and
26 backscattering measurements performed at RH<50%. The data coverage for the extensive
27 measured aerosol particle optical properties (σ_{sp} and σ_{bsp}) is generally high ranging from around
28 60% to 95%. Exception are σ_{sp} measurements in the blue (450 nm) and in the red (700 nm) and
29 σ_{bsp} measurements at CMN where the three wavelengths nephelometer was implemented starting
30 from 2014. Consequently, also SAE and g has low data coverage at CMN. Moreover, lower data
31 coverage (< 40%) was registered at PLA and VHL. The data coverage for the intensive aerosol
32 particle optical properties (SAE and g) is generally lower compared to the data coverage of σ_{sp} and



1 σ_{bsp} . This is because the intensive optical properties are calculated from hourly σ_{sp} and σ_{bsp} data
2 higher than 0.8 Mm^{-1} to avoid noise in the calculations. As a consequence, the data coverage of
3 the intensive properties is lower at those stations measuring usually low σ_{sp} and σ_{bsp} (e.g. mountain
4 and remote sites). For example, at JFJ the SAE and g data coverage is of around 54% and 22%,
5 respectively. At TRL these values are even lower, with 21% and 1%, respectively. However, as
6 reported in Table S3, at the majority of the stations the data coverage of SAE and g is higher than
7 60%.

8

9 **3. Results/Discussion**

10

11 **3.1 Variability of σ_{sp}**

12 Figure 2 shows the box-and-whiskers plots of σ_{sp} measured at the stations included in this
13 investigation. Table S4 and Figure S2 in the Supplementary Material report, respectively, the
14 statistics of σ_{sp} (mean, standard deviation, minimum and maximum values and 5th, 25th, 50th, 75th,
15 and 95th percentiles) and frequency and cumulative frequency distributions.

16 In Fig. 2, data are grouped based on their geographical location (cf. Fig. 1) and ordered based on
17 their placement, from mountain sites to urban sites. In each geographical sector, an increasing
18 gradient of σ_{sp} is generally observed when moving from mountain to regional and to urban sites.
19 Thus, σ_{sp} measured at mountain sites is always lower compared to measurements performed at
20 other placements (coastal to urban) even if exceptions are observed in some sectors. A large
21 range of σ_{sp} coefficients is observed across the network ranging from median values lower than 10
22 Mm^{-1} to values higher than 40 Mm^{-1} . The observed variation is consistent with the differences in
23 particulate matter (PM) mass concentrations, PM chemical composition and particle number
24 concentration observed across Europe as described for example by Putaud et al. (2010) and Asmi
25 et al. (2011). Figure 3 shows the relationship between the mean particle number concentration
26 measured at different stations during 2008 – 2009 reported in Asmi et al. (2011) and the mean σ_{sp}
27 measured over the same period (where available). As reported in Fig. 3, a good correlation is
28 observed between N50 (mean/median particle number between 50 nm and 500 nm) and N100
29 (mean/median particle number between 100 nm and 500 nm) and mean σ_{sp} . Overall, the lowest σ_{sp}
30 is on average measured at remote stations either because of: a) their altitude, for example JFJ
31 located in central Europe at more than 3500 m a.s.l. and CHC in Bolivia at around 5300 m a.s.l., or
32 because their large distance from pollution sources, for example the coastal ZEP and TRL
33 stations and some regional/continental sites in the Nordic and Baltic sector such as BIR, SMR and
34 PAL. The Arctic (ZEP) and Antarctic (TRL) monitoring stations are located in undisturbed
35 environments with minimal influence from the local settlement since these are located above the
36 inversion layers. The PAL station (Nordic and Baltic) is located in a remote continental area and
37 the low σ_{sp} measured at this site are mainly due to the absence of large local and regional pollution



1 sources (e.g. Aaltonen et al., 2006). Conversely, higher σ_{sp} (medians $> 40 \text{ Mm}^{-1}$) are on average
2 registered at more polluted sites such as some urban sites in southern Europe (UGR and DEM),
3 some regional sites in eastern and central Europe (KPS and IPR, respectively), and one coastal
4 site in the Nordic and Baltic sector (PLA). Finally, at all stations included in this work, the skewness
5 of σ_{sp} distributions (cf. Table S4) is higher than one and ranged between 1.4 at PLA and 10.6 at
6 TRL (skewness calculated from hourly averaged data). Positive skewness is usually observed for
7 positive defined parameters having a frequency distribution with a pronounced right tail indicating
8 the presence of high positive values. Figure S2 in the Supporting Material shows the frequency
9 and cumulative frequency distributions for σ_{sp} for each station evidencing the presence of these
10 right tails.

11

12 **3.1.1 σ_{sp} at mountain observatories**

13 Differences can be observed among stations with similar placements but different geographical
14 locations. Among the mountain stations higher mean σ_{sp} is on average measured at HPB and IZO
15 (cf. Table S4). HPB station is likely to be more influenced by the PBL than other mountain stations
16 due to its lower altitude (Nyeki et al., 2012; Collaud Coen et al., 2017), whereas IZO is largely
17 influenced by Saharan dust outbreaks transporting dust toward the station (e.g. Rodriguez et al.,
18 2011) thus increasing σ_{sp} . In fact, at IZO the median value of σ_{sp} is among the lowest measured at
19 these mountain sites (around 7 Mm^{-1} ; cf. Table S4) indicating that sporadic but extremely intense
20 pollution episodes due to Saharan mineral dust outbreaks strongly affect the mean σ_{sp} at this
21 station. The lowest median σ_{sp} at mountain sites are on average measured at JFJ probably due to
22 the higher altitude of this station compared to other mountain stations included in this work and/or
23 the distance from important pollution sources. Moreover, Collaud Coen et al. (2017) reported a low
24 PBL influence at this site due to the location of the station in a dominant position in the whole
25 mountainous massif. CHC registers higher median σ_{sp} compared to IZO or JFJ despite its location
26 at around 5300 m a.s.l. likely due to the influence of the emissions from the city of La Paz (3600 m
27 a.s.l.) located around 30 km far from CHC and the local topography which facilitates the uplift of air
28 masses toward the CHC observatory (Collaud Coen et al., 2017).

29

30 **3.1.2 σ_{sp} at regional/continental observatories**

31 Regional sites present a large variability in σ_{sp} coefficients across Europe with the lowest values
32 measured at BIR and SMR (Nordic and Baltic) and the highest at IPR (central Europe) and KPS
33 (eastern Europe). At both IPR and KPS, the frequent wintertime episodes linked to strong stable air
34 with thermal inversion strongly affect the level of pollution at these sites (e.g. Putaud et al., 2014;
35 Molnár et al., 2016). On the other side, it is known that the IPR station, even though it lies several
36 tens of kilometers away from large pollution sources, is located in an area (the Po Valley) which is



1 one of the most polluted regions in Europe (e.g. van Donkelaar et al., 2010). Among the
2 continental sites, VHL registers on average higher σ_{sp} compared to PAL and compared to BIR and
3 SMR regional sites likely because VHL is located closer to the continent and it is consequently
4 more affected by polluted continental air masses. Moreover, the emissions from densely populated
5 areas of Helsingborg and Malmö and the city of Copenhagen located 25 km to the west, 50 km to
6 the south, and 45 km to the south-east, respectively, could also explain the relatively high σ_{sp}
7 measured at VHL (Kecorius et al., 2016). The σ_{sp} values at regional level in Western Europe (OPE
8 and CBW) are on average higher compared to those measured in the Nordic and Baltic regions
9 and lower compared to those measured at regional level in south Europe (MSY).

10

11 **3.1.3 σ_{sp} at urban observatories**

12 Among the urban background sites, lower σ_{sp} are measured at MAD and SIR compared to DEM
13 and UGR. Low σ_{sp} at MAD during the period presented here (only 2014 available for MAD) could
14 be related to the reduced formation of secondary nitrate aerosols due to the limitation in the
15 availability of ammonia in this urban environment (Revuelta et al., 2014). However, it should be
16 considered that winter episodes with high secondary nitrate concentrations are not uncommon in
17 Madrid and we are presenting here only one year of measurements for this station. On the other
18 hand, secondary inorganic aerosol concentrations recorded at SIR sub-urban observatory can be
19 considered as representative of a large geographical zone, given the rather flat orography of the
20 Parisian basin. At UGR, the accumulation, mainly in winter, of fine particles from traffic, domestic
21 heating and biomass burning explains the relatively higher σ_{sp} (e.g. Lyamani et al., 2012; Titos et
22 al., 2017). Traffic emissions, high formation of secondary sulfate and organic aerosols in summer
23 together with the transport of dust from Africa are the main reasons explaining the high σ_{sp} at DEM
24 where high $PM_{2.5}$ and PM_{10} are usually measured compared to other important Mediterranean
25 cities (e.g.: Diapouli et al., 2017; Eleftheriadis et al., 2014; Karanasiou et al. 2014; Querol et al.,
26 2009).

27

28 **3.1.4 σ_{sp} at coastal observatories**

29 The PLA coastal station registered σ_{sp} values higher compared to both other Nordic and Baltic
30 stations and other coastal sites (e.g. MHD and FKL) and amongst the highest in Europe. Kecorius
31 et al. (2016) have shown that ship emissions in the Baltic Sea contribute strongly to pollution levels
32 at PLA and that up to 50% of particles arriving at PLA are generated by processes and emissions,
33 including shipping, taking place in areas upwind the station. Moreover, Asmi et al. (2011)
34 presented some similarities in particle number concentrations measured at PLA with those
35 measured at some central European sites such as IPR due to the influence from multiple source



1 areas (cf. Fig. 3). It should be noted however, that the period with available σ_{sp} measurements is
2 very short at PLA (cf. Table 1 and Figure 7) and the data coverage is also low (cf. Table S3).
3 Consequently, more measurements at this site are needed in order to confirm the σ_{sp} values
4 reported here. The other two coastal stations (MHD and FKL) register median σ_{sp} values in the
5 upper range of σ_{sp} measured across the network mostly due to the contribution of marine aerosol in
6 winter and mineral dust in summer at MHD and FKL, respectively (cf. Paragraph 3.5).

7

8 **3.2 Variability of SAE**

9 Figure 4 shows the box-and-whiskers plots of SAE calculated at the different stations. Table S5
10 and Figure S3 in the Supplementary Material report the statistics of SAE and frequency and
11 cumulative frequency distributions, respectively. It should be noted that the comparison of SAE
12 among the different stations could be slightly biased by the different particle size cuts upstream the
13 integrating nephelometers used in this work (cf. Table 1). Currently, all ACTRIS integrating
14 nephelometers measure whole air or PM_{10} . Whole air is currently measured at mountain sites
15 (BEO, CMN, JFJ, PUY, CHC) and one coastal (MHD), and two urban/suburban (UGR and SIR)
16 observatories (cf. Table 1). At some stations, the inlet was changed from whole air to PM_{10} at a
17 given time, namely at OPE, FKL, and TRL. Given the lower scattering efficiency of aerosol
18 particles larger than $10\ \mu\text{m}$, no important differences in the SAE should be expected between
19 aerosol particles sampled with whole air and PM_{10} cut-off. At other stations the inlet was changed
20 during the measurement period from a cut-off lower than $10\ \mu\text{m}$ ($1\ \mu\text{m}$ at KPS; $2.5\ \mu\text{m}$ or $5\ \mu\text{m}$ at
21 PAL, MSA and MAD) to PM_{10} . For PAL (where a median SAE of around 1.8 was measured; cf.
22 Table S5), Lihavainen et al. (2015a) assumed that the inlet changes (from PM_5 to $PM_{2.5}$ in 2005
23 and from $PM_{2.5}$ to PM_{10} , cf. Table 1) had only minor effects on scattering, because the number
24 concentration of coarse particles is very low at PAL. Similarly, KPS observatory registers among
25 the highest SAE observed in the network (median value around 2) suggesting an aerosol particle
26 size distribution dominated by fine particles. Consequently, the inlet change from PM_1 to PM_{10} at
27 KPS had probably a minor effect on SAE. Finally, two stations (MSA and MAD) changed the inlet
28 from $PM_{2.5}$ diameter cut-off to PM_{10} . For these two Southern European stations the inlet change
29 may have had an effect on SAE especially during Saharan dust outbreaks, which are however
30 sporadic events. Thus, despite the differences in the particle diameter cut-off the comparison
31 between the different stations in terms of SAE seems feasible.

32 The SAE shows a huge variability across the geographical sectors (Fig. 4). On average, the
33 highest median SAE, around 1.8 – 2.0, are observed at all central and eastern European
34 observatories (cf. Table S5). These values are quite high indicating clearly the predominance of
35 fine particles at these two geographical locations. Moreover, high $PM_{2.5}/PM_{10}$ ratios, indicative of
36 presence of small particles, are typical for rural lowland sites in central Europe (e.g. Spindler et al.,
37 2010; EMEP, 2008). Figure S3 also shows that at central and eastern sites the SAE data have
38 very similar unimodal delta-like distributions. Exceptions are CMN, JFJ and BEO mountain sites,



1 where left-tailed distributions of SAE are observed likely due to the reduced effect of fine particles
2 from the PBL in winter and an increase in the relative importance of coarse mineral dust or sea salt
3 particles as well as aged aerosols compared to lower altitude stations in the same geographical
4 sector.

5 On average, the SAE is lower at all other geographical sectors compared to central and eastern
6 Europe even though some exceptions are observed. For example, at CBW (western Europe) the
7 median SAE reaches values around 2.1. Indeed, both polluted air masses from industrialized
8 zones of the Benelux countries and clean air masses from the sea contribute to the presence of
9 aerosol particles at this site (Crumeyroille, et al., 2010). Moreover, CBW is surrounded by several
10 large cities at a distance of about 20 to 40 km from the station, which may have contributed to the
11 high SAE measured in this geographical location. Asmi et al. (2011) have also shown that
12 background particle number concentrations at CBW are much higher than for example at BIR.
13 Median SAE close to one or lower, indicative of the fact that σ_{sp} is dominated by large particles, are
14 observed at more remote sites such as MHD, IZO, ZEP, and TRL. Low SAE at MHD was already
15 reported by Vaishya et al. (2011, 2012) and justified by the frequent presence mainly in winter of
16 coarse mode sea-salt particles, since mineral dust particles can be ruled out. In fact, air masses
17 originating from dust sources are not frequent at these sites. Similarly, the low SAE observed at
18 ZEP and TRL can be associated with the presence of coarse sea-salt particles. Conversely, the
19 SAE obtained at IZO is mainly due to the frequent presence of mineral dust particles from African
20 deserts (e.g. Rodríguez et al., 2011). Very similar bi-modal frequency distributions are observed at
21 MHD and IZO showing a pronounced left peak indicating the high probability of measuring coarse
22 particles at these sites. BIR and PLA also show an enhanced left peak in the SAE frequency
23 distributions.

24 Differently from σ_{sp} , the SAE does not show any clear gradient when moving from mountain to
25 regional/urban sites. For example, at mountain sites the median SAE ranges between around 0.7
26 at IZO to values higher than two at JFJ and CMN. As reported by Zieger et al. (2012) a SAE value
27 around 2 prevails for most of the time at JFJ and can be regarded as the typical background under
28 non-dusty conditions. Thus, the SAE values at JFJ and CMN can be considered as representative
29 of central Europe free troposphere and especially in winter when the PBL emissions at these sites
30 are reduced. This high variability of SAE at mountain sites was also reported by Andrews et al.
31 (2011). Andrews et al. (2011) reported SAE values from 11 mountaintop stations worldwide
32 ranging from less than one to more than two. Moreover, Bourcier et al. (2012) have shown that at
33 mountain sites coarse particles are transported more efficiently at high altitude by higher wind
34 speed thus probably also contributing to the observed variability of SAE at mountain sites. Also at
35 coastal sites (PLA and MHD), the SAE shows large variability with higher SAE measured at PLA
36 compared to MHD confirming a higher effect of anthropogenic emissions at PLA compared to
37 MHD. Less variability in median SAE is on average observed at regional sites, with the exception
38 of OPE where a lower SAE is observed probably due to the influence of agricultural practices in the
39 vicinity. Among the urban sites, MAD registers the lowest median SAE (1.47) compared to UGR



1 (1.69) and DEM (1.60). The lower SAE at MAD could be explained, as already noted, by the
2 reduced formation of secondary inorganic aerosols during the available measurement period.
3 Moreover, resuspended dust from vehicles could also explain the lower SAE observed at MAD
4 observatory.

5

6 **3.3 Variability of g**

7 The asymmetry parameter is widely used in radiative transfer models because it provides
8 information about how much radiation is scattered back compared to the amount of radiation
9 scattered in the forward direction. Figure 5 shows the box-and-whiskers plots of g calculated at the
10 different stations. Table S6 and Figure S4 in the Supporting Material report the statistics of g and
11 frequency and cumulative frequency distributions, respectively. Given that g is calculated from BF
12 using Equation 2 (Section 2.2.3), we report in Figure S5 in the Supporting material the box-and-
13 whiskers plots of BF whereas Table S7 reports the statistics of BF. Figure 5 and Figure S5 are
14 symmetrical, thus the lower BF the higher is g . As already observed for SAE, the g varies
15 considerably among the different stations ranging between median values around 0.49 (CMN) to
16 around 0.7 (TRL). Higher g median values are in some cases observed at mountain sites
17 compared to regional or urban environments. This is the case for example for IZO compared to
18 MSY, UGR and MAD in the southwestern European sector or HPB and JFJ compared to IPR, MPZ
19 and KOS in central Europe. However, exceptions are observed for example for CMN where the
20 median g value (only 2 years available) is the lowest in the central European sector and among the
21 lowest observed in this study. On average, g values range between 0.49 to 0.64 at mountain sites
22 with a mean value of 0.58 ± 0.05 . This value is consistent with the mean value of 0.61 ± 0.05 reported
23 by Andrews et al. (2011) at the mountain sites included in their work. Figure S6 in the Supporting
24 material reports the mean SAE (ordered from low to high values in each geographical location) and
25 g at each station used in this work and the SAE- g scatter plot. Figure S6 shows that no clear
26 relationship between g and SAE can be observed. For example, TRL and MHD observatories
27 register among the highest g observed in the network which is consistent with the very low SAE
28 measured at these stations because of the frequent presence of coarse mode sea-salt particles (cf.
29 Fig. 4). However, g values similar to TRL and MHD are also observed at stations such as PLA,
30 BIR, JFJ, and DEM, which are dominated on average by fine aerosol particles (SAE similar or
31 higher than 1.5). However, there are geographical locations (e.g. Nordic and Baltic, western and
32 southwestern Europe) where SAE increases and correspondingly the g decreases from one station
33 to another indicating a shift toward finer particles. However, this is not a general rule. In fact, the
34 same relationship is not observed for example in central or eastern Europe (cf. Fig. S6).
35 Differences in the shape of the particle number size distribution, particle shape and chemical
36 composition (e.g. refractive index, RI) are factors likely contributing to explain the poor relationship
37 observed between g and SAE. The Mie theory of polydisperse spherical particles predicts that BF
38 is lower and g correspondingly higher for coarse mode aerosol particles (for which the SAE will be
39 low) compared to fine mode particles. However, some studies deploying integrating nephelometer



1 have found that BF can be higher for coarse mode aerosol particles (such as mineral dust) than for
2 fine mode aerosol particles (Carrico et al., 2003; Doherty et al., 2005). Doherty et al. (2005)
3 suggested that an under-correction for the σ_{sp} truncation of the forward-scattered radiation (which
4 is relatively larger for coarse particles) could bias the calculated BF high. Moreover, the shape of
5 particle number size distribution is another factor affecting BF and SAE. Thus, differences in the
6 relative fractions of the fine and coarse modes could also drive the BF-SAE relationship. In fact,
7 the SAE is most sensitive to the presence of coarse mode aerosol particles compared to BF which
8 is most sensitive to small accumulation mode particles (Delene and Ogren, 2002; Collaud Coen et
9 al., 2007). Thus, depending on the shape of the particle number size distribution, BF and SAE
10 might or might not correlate. Moreover, the refractive index (RI), which is strongly related to the
11 chemical composition of the particles, is another important variable, that can affect g (e.g. Marshall
12 et al., 1995). In the work from Hansen and Travis (1974; Fig. 12) the authors showed that for a
13 given particle diameter the g parameter did non linearly decreased with increasing real RI. Thus,
14 coarse mode particles with a given RI could have an asymmetry parameter similar or lower to that
15 of fine particles with lower RI. Recently, Obiso et al. (2017) confirmed the findings by Hansen and
16 Travis (1974) showing also that a perturbation in RI of 20% has a higher effect on g compared to
17 similar relative perturbation of particle shape. On the other side, Obiso et al. (2017) showed that a
18 variation of RI for coarse particles can have a small effect on the mass scattering efficiency of the
19 particle and its spectral dependence and consequently on SAE.

20

21 **3.4 Relationships between σ_{sp} and intensive optical properties**

22 Figure 6 shows the relationships between σ_{sp} and SAE and between σ_{sp} and g at each station.
23 Mean SAE and g are calculated for each σ_{sp} bin and the bin size at each station is calculated
24 following the Freedman – Diaconis rule:

25

$$26 \text{ Bin size} = 2 \frac{\text{IQR}(x)}{\sqrt[3]{n}} \quad (\text{Eq. 3})$$

27

28 where IQR(x) is the interquartile range of the data and n is the number of observations in the
29 sample x . This kind of graphs helps in understanding which aerosol type on average dominates the
30 particle light scattering, depending on the amount of scattering measured. It should be noted that in
31 Figure 6 the number of samples available at each station is not evenly distributed among the
32 considered bins. Figure S7 in the Supplementary Material shows for some stations the SAE- σ_{sp}
33 pairs colored by the number of samples in each bin to highlight how samples are distributed among
34 the bins.

35

36 **3.4.1 g - σ_{sp} relationships**

37 The asymmetry parameter g shows the lowest values under very low σ_{sp} suggesting the
38 predominance of small fine mode particles. Andrews et al. (2011) reported similar g - σ_{sp}



1 relationships at different mountain sites and suggested that the removal of large particles by cloud
2 scavenging or by deposition during transport could explain the observed low g under a clean
3 atmosphere. They also suggested that the formation of new particles followed by
4 condensation/coagulation could generate small but optically active particles. Here, we show that
5 this behavior of BF or g as a function of σ_{sp} was observed at all sites, not only at mountain sites.

6 The parameter g then increases with increasing σ_{sp} indicating a shift of the particle number size
7 distribution towards the larger end of the accumulation mode. Delene and Ogren (2002), Andrews
8 et al. (2011) and Pandolfi et al. (2014) showed that BF tends to decrease with increasing aerosol
9 loading, consistent with the observed increase of g . For comparison with previous works, Figure S8
10 in the Supplementary Material shows the BF- σ_{sp} relationships for all observatories evidencing the
11 aforementioned BF decrease with increasing σ_{sp} .

12 The shift of the particle number size distribution towards the large end of the fine mode with
13 increasing σ_{sp} is probably the main reason causing the observed increase of g (and the decrease
14 of BF, cf. Fig. S8). A possible explanation for this shift could be a progressive aging of atmospheric
15 aerosol particles. Then, at the majority of stations, the variation of g is less pronounced under
16 periods of high particle mass concentrations suggesting changes mostly in the coarse aerosol
17 particles mode rather than in the fine mode.

18

19 **3.4.1 SAE- σ_{sp} relationships**

20 As reported in Figure 6, at some stations the SAE progressively increases with σ_{sp} in the σ_{sp} range
21 where the g parameter increases as well. The increase of both g and SAE with σ_{sp} , observed for
22 example at the Nordic and Baltic, central and eastern European observatories, could be related to
23 the different effects that different particle sizes have on SAE and g . A progressive increase of SAE
24 with σ_{sp} would suggest an increasing relative importance of fine aerosol particles. The origin of
25 these fine particles is probably different depending on the location of the measuring site. For the
26 remote PAL site, for example, Lihavainen et al. (2015b) observed an increase of both σ_{sp} and SAE
27 with increasing temperature due to increasing formation of BSOA (biogenic secondary organic
28 aerosols) with increasing ambient temperatures, thus likely driving the σ_{sp} -SAE relationships
29 reported in Fig. 6 for PAL. The BSOA from gas-to-particle formation over regions substantially
30 lacking in anthropogenic aerosol sources such as the European boreal region (Tunved et al., 2006)
31 are probably strongly contributing to the σ_{sp} -SAE relationships observed at other Nordic and Baltic
32 sites such as SMR. At polluted sites such as those located in central and eastern Europe the
33 anthropogenic aerosol emission and the active secondary aerosol production in the region (e.g. Ma
34 et al., 2014) are probably driven the σ_{sp} -SAE relationships reported in Fig. 6. For higher σ_{sp} , the
35 σ_{sp} -SAE relationships changed and a progressive shift toward relatively larger particles is on
36 average observed with increasing σ_{sp} . However, at the majority of northwestern, central and
37 eastern European stations, the SAE keeps values around or higher 1.5 under high particle load
38 indicating that high σ_{sp} is dominated by fine particles. An exception is MHD where SAE increases



1 with increasing σ_{sp} keeping values on average lower than 1.4 under high particle load (cf. Fig. 6).
2 As already observed, low SAE at MHD is mainly due to the predominance of sea-salt coarse
3 particles at this site (Vaishya et al., 2011). Conversely, at some sites in South Europe (e.g. MSA,
4 MSY, IZO, DEM) the SAE reaches values around one or lower under high particle load indicating
5 that at these stations high σ_{sp} is dominated by mineral dust coarse particles mainly from African
6 deserts. Exceptions are two urban sites in Southwestern Europe (UGR and MAD) where fine
7 particle likely mostly from traffic (and also from biomass burning at UGR) on average dominate the
8 highest measured σ_{sp} . Similar σ_{sp} -SAE relationships, as those reported in Fig. 6, were observed by
9 Andrews et al. (2011) at mountain sites and by Delene and Ogren (2002) at marine sites. Among
10 the lowest SAE are observed at IZO, the station closest to the African continent. Interestingly, at
11 IZO the SAE shows the highest gradient for σ_{sp} coefficients in the range of 0-50 Mm^{-1} whereas the
12 gradient is much lower for σ_{sp} higher than 50 Mm^{-1} being the SAE almost constant for σ_{sp} higher
13 than 100 Mm^{-1} . IZO station is often in the free troposphere and high loading at this station are only
14 registered under Saharan dust events, thus almost only mineral dust is measured at IZO. Normally
15 the long-rang transport mineral dust particle don't have a significant fraction above 10 μm because
16 of the short lifetime, thus likely explaining the constant SAE observed at IZO under high aerosol
17 loading.

18
19

20 **3.5 Seasonal variability**

21 Figures 7, 8 and 9 present the annual cycles of σ_{sp} , SAE and g , respectively, at each site. Overall,
22 strong seasonal cycles of σ_{sp} and intensive aerosol particle optical parameters are observed at the
23 majority of the stations even if exceptions are observed. Given the important role that the station
24 placement plays in the seasonal cycles of aerosol parameters, the analysis is presented below
25 separately for mountain observatories and for low altitude observatories.

26

27 **3.5.1 Seasonal variability at mountain observatories**

28 At the mountain stations (PUY, HPB, JFJ, CMN, BEO, MSA, and IZO), σ_{sp} peaks in spring/summer
29 whereas lower σ_{sp} values are measured in autumn/winter. Similar findings were for example
30 already reported by Nyeki et al. (1998) for JFJ and summarized by Andrews et al. (2011) for many
31 mountain top stations worldwide and by Pandolfi et al. (2014) for MSA station. Different factors
32 contribute to the σ_{sp} increase in spring/summer at the mountaintop observatories, such as the
33 increase of the boundary layer height and stronger upslope winds during the warmest months.
34 Moreover, specific events such as Saharan mineral dust outbreaks, may contribute to the
35 increased σ_{sp} observed at mountain stations in spring/summer, and especially in southern Europe
36 (e.g. Pey et al., 2013; Pandolfi et al., 2014; Rodríguez et al., 2011). At IZO, σ_{sp} peaks strongly in
37 July-August because of the very high influence of African mineral dust at this station during these



1 months (e.g. Alastuey et al., 2005; Diaz et al., 2006). At the mountaintop CHC observatory, σ_{sp}
2 progressively increases during the dry season, from May to October, reaching lower values during
3 the rainy season (from December to April). Moreover, during the dry season the new particle
4 formation events, taking place at CHC with one of the highest frequency reported in the literature
5 so far (Rose et al. 2015), can introduce very small particles that grow to the nucleation and Aitken
6 mode. At the mountain stations, both SAE and σ_{sp} are on average higher in summer compared to
7 the winter period, thus suggesting a higher anthropogenic influence at these sites during the
8 warmest months. The summer SAE increase is more evident at some mountain stations, e.g. HPB,
9 CMN, and BEO, compared to other mountain stations such as JFJ and MSA. Less pronounced
10 SAE seasonal variation at JFJ was related by Bukowiecki et al. (2016) to the rather constant
11 composition of the JFJ aerosol. At the southern station of MSA the observed less pronounced
12 seasonal cycle of SAE could be related with the Saharan dust outbreaks which contrast the PBL
13 transport of fine particles observed at other mountain sites. At IZO, the SAE reaches the lowest
14 values during July-August being the Saharan dust outbreaks very intense at this site during this
15 period.

16 Overall, the g parameter shows opposite seasonal cycles compared to SAE at almost all mountain
17 stations with the exception of JFJ and BEO where g slightly increases with SAE in summer. At
18 almost all mountain stations, the seasonal variations of SAE and g are less pronounced compared
19 to the seasonal variation of σ_{sp} indicating larger seasonal variation in the extensive aerosol optical
20 properties than in the intensive properties. For example, the median σ_{sp} values at MSA increase by
21 around 800% during summer (JJA) compared to winter (DJF), whereas SAE and g increase by
22 around 5-7%. Similar relative increases are observed at JFJ (660%, 16% and 11% for σ_{sp} , SAE
23 and g , respectively) whereas the relative increases are much higher at BEO, especially for σ_{sp}
24 (around 1300%) and SAE (26%). At CMN, the median σ_{sp} value increases by around 400% from
25 winter to summer, whereas SAE and g increase and decrease, respectively, by around 46% and
26 6%, respectively. At CHC, the SAE decreases as the σ_{sp} increases moving from wet to dry season,
27 indicating an increasing effect of coarse particles on σ_{sp} during the dry season. At PUY, σ_{sp} peaks
28 from March to September and this increase is accompanied by a small SAE increase. Venzac et
29 al. (2009) and Boulon et al. (2011) have shown that PUY is more often influenced by the free
30 troposphere or residual layers in winter and spring compared to the summer season.

31

32 **3.5.1 Seasonal variability at low altitude observatories**

33 At some of the low altitude observatories, the seasonal variation of particle scattering is opposite
34 compared to the variations observed at mountain sites, σ_{sp} being higher in winter and lower in
35 summer. MHD, CBW and SIR in the western sector, IPR, MPZ and KOS in central, KPS in eastern
36 and UGR in south-western Europe show such increase in particle mass concentration in
37 wintertime. The reasons causing these marked seasonal cycles are probably different depending
38 on the geographical sector considered.



1 **3.5.1.1 Central and eastern Europe**

2 Central and eastern European observatories show marked seasonal cycles of both extensive and
3 intensive aerosol particles optical properties. In these regions, less horizontal and vertical pollutant
4 dispersion in winter, due to a higher frequency of stagnant conditions and temperature inversions,
5 play an important role in accumulating aerosols. As a consequence, as reported in Figure 7, the σ_{sp}
6 is much higher in winter compared to summer. SAE and g also show marked season cycles in
7 these regions, being the SAE (g) higher (lower) in summer compared to winter (cf. Fig. 8). Ma et al.
8 (2014) have shown that at MPZ an increased SAE in summer is mainly explained by the variation
9 of the particle number size distribution. Thus, high concentrations in spring and summer of small
10 particles during new particle formation and subsequent growth cause the observed increase of
11 SAE during warmest months.

12

13 **3.5.1.2 Nordic and Baltic regions**

14 At the Nordic and Baltic sites, the monthly variation of σ_{sp} is on average less pronounced
15 compared to the central or eastern European stations and especially at BIR, SMR and PAL. This is
16 likely due to the placement of these stations located in remote areas with different meteorology
17 (e.g. less pronounced PBL variations) and where on average much lower σ_{sp} values are measured
18 compared to other European sites. Moreover, this could also indicate the importance of
19 anthropogenic sources like domestic heating in central and eastern Europe in winter. However, the
20 monthly variation of SAE and g is rather pronounced at these Nordic and Baltic observatories: SAE
21 (g) increases (decreases) in summer compared to winter indicating the predominance of relatively
22 smaller particles during the warmest months. Similar findings were reported for the SMR and PAL
23 observatories by Virkkula et al. (2011) and Lihavainen et al. (2015a), respectively. The observed
24 seasonal variations in intensive aerosol optical properties were related to both the transport of
25 different air masses at these remote sites depending on the season and the enhanced formation of
26 BSOA in summer (e.g. Lihavainen et al., 2015a). Lihavainen et al. (2015a) and Virkkula et al.
27 (2011) also reported a lower single scattering albedo in winter compared to summer at PAL and
28 SMR, respectively, frequently dropping below 0.7 at SMR due to a significant contribution from light
29 absorbing carbon, mostly from residential wood combustion. Thus, they have shown that aerosol
30 particles observed in summer at SMR and PAL had the potential to cool the atmosphere more
31 efficiently than those observed during winter. Similar intensive optical properties season cycles
32 were observed at BIR.

33

34 **3.5.1.3 Western Europe**

35 Similarly to the Nordic and Baltic regions, differences in aerosol sources and sinks are the likely
36 reasons explaining the seasonal variation of σ_{sp} , SAE and g observed in western Europe. Marked
37 σ_{sp} seasonal cycles are observed at all low altitude western European observatories, with higher
38 values measured in winter compared to summer. On average, at these sites, SAE (g) is higher
39 (lower) in summer compared to winter. O'Connor et al. (2008) and Vaishya et al. (2011, 2012)



1 showed that the background marine aerosol measured at MHD contains a strong and significant
2 seasonal cycle with sea-salt dominating in winter and biogenic organic aerosol dominating the
3 submicron sizes in summer. This is consistent with the observed season cycles of SAE and *g*
4 reported here for MHD.

5

6 **3.5.1.4 South Europe**

7 Among the southern European observatories, marked seasonal variation for σ_{sp} is observed
8 especially at UGR, MSY and FKL. At the urban UGR site, the mean aerosol type is very different in
9 winter compared to summer. As evidenced by the seasonal cycles of SAE and *g*, aerosol particles
10 are generally finer in winter at UGR compared to the summer season as already observed for
11 example by Lyamani et al. (2010; 2012) and Titos et al. (2012). This is likely due to the
12 accumulation of fine particles, mainly from traffic, domestic heating and biomass burning, favored
13 by stagnant conditions and atmospheric inversions during winter. In summer, the higher frequency
14 of Saharan mineral dust outbreaks at this site increases the mean size of the particles during the
15 warmest months. At the MSY regional site, the higher efficiency of the sea breeze in transporting
16 pollutants from the urbanized/industrialized coastline toward regional inland areas during the
17 warmer season mainly explains the summer increase in aerosol particle mass concentration
18 observed at this site (e.g. Pandolfi et al., 2011). Moreover, the enhanced formation of secondary
19 sulfate and organic matter in summer together with frequent Saharan mineral dust outbreaks,
20 strongly contribute to the observed seasonal cycle for σ_{sp} and intensive properties at MSY site. The
21 σ_{sp} peak observed at MSY in March is due to the winter pollution episodes typical of the western
22 Mediterranean basin (WMB) (e.g. Pandolfi et al., 2014a and references therein). During these
23 episodes, the accumulation of pollutants close to the emission sources is favored by anticyclonic
24 conditions coupled with strong atmospheric inversions. During such conditions, pollutants
25 accumulate in the PBL and can subsequently reach the station when PBL height increases. On
26 average, at MSY low SAE are measured in April and October likely due to the occurrence of
27 Saharan dust outbreaks during these months. At FKL no intensive optical aerosol properties are
28 available. The high σ_{sp} in summer at this site is also associated with mineral dust storm events as
29 for example reported by Vrekoussis et al. (2005). However, mineral dust storms in the
30 Mediterranean are not the only reason for the observed increased σ_{sp} in summer. In fact, as for
31 example reported by Kalivitis et al. (2011) for FKL and Pandolfi et al. (2011) for MSY, ammonium
32 sulfate and particulate organic matter, whose concentrations increase in summer in the
33 Mediterranean Basin, were assumed as important contributors to σ_{sp} during the warm season. At
34 the DEM urban observatories, the high σ_{sp} measured in spring are linked to Saharan dust
35 outbreaks as also supported by the seasonal cycles of SAE and *g* which showed the lowest and
36 highest, respectively, values in spring.

37

38

39



1 3.6 Trends

2 Trends of σ_{sp} , SAE and BF are studied for those stations having more than 8 years of data (13
3 observatories). Generally, it is recommended to have more than 10 years of data for trend studies.
4 Among the ACTRIS stations, PAL, SMR, MHD, HPB, IPR, JFJ, and UGR have more than 10 yr of
5 data, whereas at PUY, MPZ, CMN, BEO, KPS, and IZO, 8 or 9 years are available. These stations
6 are included in order to improve the spatial coverage, similarly as in Collaud Coen et al. (2013).
7 The Theil Sen statistical estimator (Theil, 1950; Sen, 1968) is used here to determine the
8 regression parameters of the data trends, including slope, uncertainty in the slope and p-value.
9 The Theil Sen method provides similar results as the Mann-Kendall test and it is implemented for
10 example in the Openair Package available for R space (Carslaw, 2012; Carslaw and Ropkins,
11 2012). The applied method yields accurate confidence intervals even with non-normal data and it is
12 less sensitive to outliers and missing values (Hollander and Wolfe, 1999). Monthly means are used
13 for trend analysis and the data are deseasonalized. The data coverage of σ_{sp} is higher than 70% at
14 all stations included in trend analyses with the exception of IZO where the σ_{sp} data coverage is
15 55%. For SAE, the data coverage is higher than 65% at all sites with the exception of PAL (54%),
16 PUY (59%), and IZO (52%). For BF, the data coverage is higher than 65% with the exception of
17 PAL (26%), PUY (43%), BEO (47%) and IZO (27%). At the remote (PAL) or mountain stations
18 (PUY, BEO, and IZO), the percentage for the intensive aerosol particle optical properties is lower
19 because of a higher probability of measuring σ_{sp} lower than the threshold (0.8 Mm^{-1}) selected for
20 the calculation of SAE and BF. Table 2 reports the trends observed for σ_{sp} , SAE and BF at the
21 thirteen observatories included in this analysis. Magnitude and statistical significance of the trends
22 for these parameters are reported in Table S8 in the Supporting Material. It should be noted that
23 changes in particle size cut-off reported for PAL and KPS (cf. Table 1) may have affected the
24 reported trend analyses at these stations, but estimating the impact of these changes in the
25 observed trend is not simple. However, as already noted, Lihavainen et al. (2015a) reported that at
26 PAL the inlet changes had minor effects on scattering, because the number concentration of
27 coarse particles is very low at this observatory. KPS is dominated by very fine particles and the
28 change from PM_1 to PM_{10} had probably a minor effect on σ_{sp} , SAE and BF. Moreover, at KPS the
29 inlet was changed in April 2008, less 1.5 years after the beginning of the measurements thus likely
30 having a minor effect in the trend analysis performed at this site over the period 2006 – 2014. The
31 FKL observatory was removed from trend analysis because the inlet was changed from whole air
32 to PM_{10} in 2009, from PM_{10} to PM_1 in 2011, and again from PM_1 to PM_{10} in 2013 (cf. Table 1), thus
33 likely having a major effect on the measured particle optical properties.

34 In Table 2, a comparison with previous trends analysis results presented by Collaud Coen et al.
35 (2013) for aerosol particle optical properties and by Asmi et al. (2013) for particle number
36 concentrations is also reported.

37



1 **3.6.1 Trends of σ_{sp}**

2 Overall, σ_{sp} decreases at the majority of the stations included in this work. Significantly decreasing
3 trends for σ_{sp} are observed at: the two Nordic and Baltic observatories (PAL for the period 2000 –
4 2010 and SMR); at two observatories (HPB and IPR) out of five observatories in central Europe;
5 and at the two observatories in southwestern Europe (IZO and UGR). The trends are not
6 statistically significant in western (MHD and PUY) and eastern (BEO and KPS) Europe. The
7 highest magnitude of σ_{sp} trend [Mm^{-1}/yr] (cf. Table S8 in the Supplementary Material) is observed
8 at the polluted IPR observatory. Conversely, the lowest magnitude is observed at the remote PAL
9 observatory. For the periods considered in this work, the total reductions (TR) for σ_{sp} range
10 between around 30% (SMR) and 60% (IZO). The high TR observed at IZO might be affected by
11 the intensity and frequency of Saharan dust outbreaks at this site. However, estimating the effects
12 of these events at IZO is beyond the scope of this study. Overall, the observed decreasing trends
13 of σ_{sp} are consistent with the uniform decrease in aerosol optical depth observed in Europe
14 (AERONET data in Li et al., 2014). A statistically significant decreasing trend of σ_{sp} at IPR was also
15 reported by Putaud et al. (2014) for the period 2002 – 2010. As reported in Table 2 statistically
16 significant decreasing trend for σ_{sp} is observed at around 50% of the stations considered here.
17 Overall, the observed statistically significant decreasing trends of σ_{sp} are consistent with the
18 demonstrated reduction of PM concentration in the atmosphere in Europe in these last decades
19 thanks to the implementation of European/national/regional/local mitigation strategies. These
20 decreasing trends are also consistent with trends of aerosol chemistry derived from observations in
21 urban environments in Europe (e.g. EEA, 2013; Barmpadimos et al., 2011; Titos et al., 2014;
22 Pandolfi et al., 2016), regional and remote environments in western Mediterranean (Cusack et al.,
23 2012; Pandolfi et al., 2016) and in general with derivation of trends for aerosol chemistry across
24 Europe (Tørseth et al., 2012). Recently, Collaud Coen et al. (2013) showed that trends in σ_{sp} are
25 observed at most of the US continental sites and that these trends are generally consistent with the
26 strong SO_2 and PM reductions observed in the US (Asmi et al., 2013; EPA, 2011). Conversely, in
27 Europe the strong decreasing trend observed for SO_2 (e.g. Tørseth et al., 2012; Henschel et al.,
28 2013) and, with a lower spatial homogeneity and statistical significance, for $\text{PM}_{2.5}$ (e.g. EEA, 2016)
29 is not observed for aerosol optical properties. As reported in Collaud Coen et al. (2013) the
30 reasons why at some of the European sites no significant trends are observed, might be related to
31 the spatial inhomogeneities and under-representation of continental Europe PBL sites (e.g. Laj et
32 al., 2009) and/or the timing of the SO_2 and PM trends for the US and Europe. In Europe the
33 emissions reductions were greater for the period 1980–2000 compared to the period 2000 – 2010
34 (e.g. Colette et al., 2016; Tørseth et al., 2012; Manktelow et al., 2007), thus the measurements of
35 optical particle properties in Europe may not go back far enough to reflect the time period with the
36 largest emission reductions. Tørseth et al. (2012) reported average reductions for ambient sulfate
37 and nitrate mass concentrations in Europe of -12% and -1%, respectively, during 2000 – 2009
38 compared to -24% and -7%, respectively, during 1990 – 2000. They also reported statistically



1 significant decreases of PM_{10} and $PM_{2.5}$ mass concentrations at around 50% of European sites
2 with total reductions of -18% and -27%, for PM_{10} (24 sites) and $PM_{2.5}$ (13 sites), respectively,
3 during 2000 – 2009. A direct comparison between the stations included in this work and those
4 included in Tørseth et al. (2012) is not possible because of the different timing of reported σ_{sp} and
5 PM mass concentration measurements. At those stations where a significant decreasing trend for
6 σ_{sp} is observed and considering a period of 10 yr (even if not coincident for all stations), the total
7 reduction for σ_{sp} in Europe is around -35% (cf. Table S8) consistent with the trend reported by
8 Tørseth et al. (2012) for PM in Europe. Quite good agreement, even though again likely biased by
9 the different timings, is also observed comparing PM mass concentration and σ_{sp} trends by
10 geographical sectors. A significant total reduction around -40 ÷ -30% was reported for PM_{10} and
11 $PM_{2.5}$ in the Nordic and Baltic sector by Tørseth et al. (2012; cf. Fig. 7 in Tørseth et al. (2012)) in
12 close agreement with the statistically significant total decrease of σ_{sp} around -34% reported for PAL
13 during 2000 – 2010 (cf. Table S8). In the Western sector (MHD) the decreasing trend for $PM_{2.5}$
14 during 2000 – 2009 was insignificant (-10 ÷ 0%) as reported here for σ_{sp} during the period 2001 –
15 2010. In the Central sector statistically significant decreases for $PM_{2.5}$ and PM_{10} mass
16 concentrations ranged between -20% and -40% during a 10 yr period (2000 – 2009) and the total
17 reduction for σ_{sp} ranged between -38% (HPB) and around -48% (IPR). In the Southwestern
18 European sector the total reduction for σ_{sp} is around -32% (at UGR) and -60% (at IZO), whereas
19 Tørseth et al. (2012) reported around -20 ÷ -40% decrease for the PM_{10} mass concentration. To
20 further confirm the observed close agreement between PM trends reported in literature and the
21 trends of σ_{sp} in this work, Table S9 in the Supporting Material reports the comparison between σ_{sp}
22 and PM_{10} and/or $PM_{2.5}$ mass concentration trends calculated at those stations where simultaneous
23 σ_{sp} and PM mass concentration measurements are available. As reported in Table S9 both the
24 observed total reductions and the statistical significance of the trends are very similar for σ_{sp} and
25 PM_{10} .

26

27 3.6.2 Trends of SAE and BF

28 The trends for SAE are estimated for three different quantities, namely: the SAE calculated as
29 linear fit using three wavelengths (b-g-r), using the blue and the green wavelengths (b-g) and using
30 the green and red wavelengths (g-r). For the periods considered in this work (in bold in Table 2),
31 the SAE calculated using the three wavelengths (b-g-r) shows statistically significant trends at five
32 sites. At PAL (Nordic and Baltic), PUY (western Europe) and BEO (eastern Europe) decreasing
33 trends are observed, whereas increasing trends are observed at HPB (central Europe) and UGR
34 (southwestern Europe). Uniform negative trends of columnar Ångström exponent from AERONET
35 data were reported by Li et al. (2014) across Europe and these trends were ascribed to reduced
36 fine-mode anthropogenic emission. The positive SAE trend observed at HPB and UGR would
37 suggest a shift of the accumulation mode particles towards smaller sizes and/or a change in the



1 coarse aerosol mode. For example, the SAE increase at UGR could be probably explained by a
2 progressive relative importance of fine particles emissions driven by a progressive reduction of
3 coarse particles for example from construction/demolition works due to the economic crisis which
4 affected Spain from 2008 (e.g. Lyamani et al., 2011; Querol et al., 2014; Pandolfi et al., 2016). In
5 fact, Titos et al. (2014) reported statistically significant decreasing trend for PM₁₀ fraction during the
6 period 2006 – 2010 whereas no trend was observed for PM₁ fraction. Moreover, at UGR,
7 statistically significant increasing trend is also observed for the SAE calculated using the green and
8 red wavelengths (g-r), likely more sensitive to the coarser particle mode, whereas the trend was
9 non-statistically significant for the SAE b-g. The possible change in the coarse aerosol mode at
10 UGR is likely also causing the observed statistically significant increasing trend of BF (cf. Table 2),
11 given that a positive trend of BF would be consistent with a shift of the accumulation mode
12 particles towards smaller sizes. Similarly, statistically significant increasing trends for both SAE and
13 BF are also observed at SMR (SAE b-g) and HPB. Statistically significant increasing trends of BF
14 are also observed at the other Nordic and Baltic stations (PAL) and at PUY (western Europe),
15 where the SAE shows statistically significant decreasing trends, and at IPR (central Europe) where
16 the trend of SAE was insignificant. Thus, overall, the trends of BF are positive at all stations where
17 BF measurements are available. The opposite sign of the trends of SAE and BF at PAL and PUY
18 could be due to different effects that different particle sizes have on SAE and *g* or a progressive
19 change in the mean diameter of the fine mode aerosol. Further research involving for example size
20 distribution data and Mie calculation could help in understanding the differences observed in some
21 cases between SAE and BF (or *g*). Recently, Korras-Carraca et al. (2015) have shown that the
22 column integrated *g* from Modis-Terra had widely statistically significant positive trends (2002-
23 2010) with stronger increases observed in the eastern and southern Black Sea, as well as over the
24 Baltic and Barents seas. Moreover, both Modis-Terra and Modis-Aqua produce positive trends of *g*
25 in the eastern Mediterranean Sea and the eastern coast of the Iberian Peninsula. Positive trends
26 for *g* would correspond to negative trends for BF. The difference observed with our work could be
27 due to the different variability often observed between near-surface measurements and column
28 integrated measurements which can confound the relationship between surface and column optical
29 properties (e.g. Bergin et al., 2000; Lyamani et al., 2010). Although, it was shown that mid altitude
30 station might be globally representative of the whole atmospheric column (Chauvigne et al., 2016).

31

32 3.6.3 Comparison with previous trend analyses

33 Table 2 shows the comparison, over the same periods, between the trend analyses performed in
34 this work and the analyses presented by Collaud Coen et al. (2013) for aerosol particle optical
35 properties and by Asmi et al. (2013) for particle number concentrations (N_{LDL-500}, N₂₀₋₅₀₀ and N<sub>100-
36 500</sub>). An agreement with the results from Collaud Coen et al. (2013) is observed for JFJ where
37 consistent insignificant trends are detected for the three periods reported in Collaud Coen et al.
38 (2013). For MHD, we calculated a non-significant increasing trend for σ_{sp} during 2001 – 2010,



1 whereas Collaud Coen et al. (2013) reported a statistically significant increasing trend for the same
2 period. At PAL, non-statistically significant trend for σ_{sp} is observed here and in Collaud-Coen et al.
3 (2013) for the period 2001 – 2010, whereas we observe a statistically significant decreasing trend
4 for the period 2000 – 2010. Moreover, at PAL, we observe statistically significant decreasing trend
5 for SAE during the two common periods which were insignificant in Collaud Coen et al. (2013). It
6 should be noted that Collaud Coen et al. (2013) reported insignificant SAE trend at PAL using the
7 Mann Kendall test whereas they reported statistically significant decreasing trends using the
8 GLS/ARB and LMS methods, consistent with our work. These differences are thus likely due to the
9 relative short period used in these trend analyses and the different sensitivity of the methods used
10 to the presence of missing values or outliers especially at PAL where σ_{sp} is very low (cf. Fig. 2). For
11 example, in this work the SAE calculated for PAL during the year 2007 was removed from the
12 trend analysis due to the presence of too many extreme high SAE values, thus also likely
13 explaining the difference observed for SAE with the work from Collaud Coen et al. (2013).
14 Moreover, here we use de-seasonalized monthly means for trend analyses whereas Collaud-Coen
15 et al. (2013) used de-seasonalized medians with different time granularity (3 days) thus likely
16 affecting the comparison, especially over relatively short periods.

17 A comparison of trends analysis results between σ_{sp} and the particle number concentration is not
18 straightforward as the σ_{sp} measurements are more sensitive to the particle number concentration in
19 the upper end of the fine mode than to smaller particles. For example, Asmi et al. (2013) reported
20 that, globally, no strong similarities were observed between σ_{sp} and N trends and that the N trends
21 are controlled by particles in the larger range of the Aitken mode and smaller range of the
22 accumulation mode, e.g. ca. 50–150 nm diameter. In this work, as reported in Table 2, the
23 statistically significant decreasing trend reported for N during the period 2001 – 2010 is not
24 observed for σ_{sp} . However, differences are also observed at PAL between N20 and N100 mainly
25 because DMPS measurements at PAL had long gaps during periods with unusually low
26 concentrations thus effectively removing low concentrations from the trend analysis (Asmi et al.,
27 2013).

28

29 **3.6.4 Daytime and nighttime trend analyses at mountain sites**

30 Finally, the analysis of the trends during daytime (08:00 – 16:00 GMT) and nighttime (21:00 –
31 05:00 GMT) by season at mountain stations are also analyzed (Table 3). This analysis could
32 provide information about changes in σ_{sp} when the mountain stations are likely affected by the PBL
33 (e.g. daytime and/or summer) or by the residual layer (e.g. nighttime in summer) or when these are
34 representative of the free troposphere (e.g. nighttime in winter). Consistently with what reported in
35 Table 2 for σ_{sp} , the trends are insignificant at JFJ, PUY CMN, and BEO irrespective of the time of
36 the day or season. The decreasing trends observed at HPB, also reported in Table 2, are



1 statistically significant only during autumn, irrespective of the time of the day. Conversely, the trend
2 observed for σ_{sp} at IZO reported in Table 2, is not observed by splitting the analysis by time of the
3 day and/or season.

4

5 **Conclusions**

6

7 This investigation presented the near-surface in-situ σ_{sp} (aerosol particle light scattering), SAE
8 (Scattering Ångström exponent), BF (backscatter fraction), and g (asymmetry parameter)
9 measurements obtained over the past decade at 28 measuring atmospheric observatories which
10 are part of the ACTRIS Research Infrastructure and most of them belong to the GAW network.
11 Results show a large variability of both extensive and intensive aerosol particle optical properties
12 across the network, which is consistent with the previously reported variability observed for other
13 aerosol particle properties such as particle mass concentration, particle number concentration and
14 chemical composition. Main findings can be summarized as follows:

15

- 16 - Overall, the highest σ_{sp} are measured at low altitude observatories in central and eastern
17 Europe and at some urban sites in south Europe whereas, the lowest σ_{sp} are observed at some
18 mountain stations and at two Arctic and Antarctic sites. Low σ_{sp} levels, comparable with those
19 measured at mountain sites, are also observed at the majority of the regional/continental
20 Nordic and Baltic observatories. The σ_{sp} values in Western Europe are on average higher
21 compared to those measured in the Nordic and Baltic regions and lower compared to those
22 measured at regional level in south Europe. Some exceptions to these general features are
23 however observed.
- 24 - In central and eastern Europe, independently from the station placement, the SAE (g) is among
25 the highest (lowest) observed across the network indicating a large predominance of fine
26 particles. In these regions, the SAE (g) is even higher (lower) in summer compared to winter
27 suggesting the shift toward the small end of the aerosol particle size distribution likely linked to
28 new particle formation events during the warmest months. On average SAE (g) is lower
29 (higher) in the Nordic and Baltic, western and southern sectors compared to central and
30 eastern Europe.
- 31 - Seasonal cycles for σ_{sp} are observed in all geographical sectors. These are especially marked
32 at regional level in central and eastern Europe where wintertime episodes linked with stable air
33 and thermal inversions favor the accumulation of pollutants. Clear annual cycles are also
34 observed at mountain sites where σ_{sp} is higher in summer because of the enhanced boundary
35 layer influence. In some cases, SAE (g) is also high (low) in summer at mountain sites
36 indicating a higher PBL anthropogenic influence during the warmer months. In the Nordic and
37 Baltic regions, the seasonal variation of σ_{sp} is less pronounced compared to central and
38 eastern Europe likely due to different meteorology and less pronounced PBL variations.
39 Despite the relatively small σ_{sp} seasonal cycles in the Nordic and Baltic regions, SAE (g)



1 increases (decreases) in these regions in summer compared to the winter period likely due to a
2 seasonal-dependent transport of air masses at these remote sites and an enhanced formation
3 of secondary organic aerosols previously observed at these sites during the warmest months.
4 At coastal sites in northwestern Europe, the presence of sea-salt particles in winter also
5 contributes to the observed pronounced seasonal cycles of SAE and g .

6 - The analysis of the systematic variability of SAE and g as a function aerosol loading (σ_{sp})
7 reveals some common patterns. At all stations, g shows the lowest values under very low σ_{sp}
8 likely because the formation of new particles in a clean atmosphere followed by
9 condensation/coagulation with consequence generation of small but optically active particles.
10 The g then sharply increases with increasing σ_{sp} indicating the shift of the particle number size
11 distribution toward the larger and of the accumulation mode. Then, under periods of high
12 particle mass concentrations, the variation of g is less pronounced at the majority of the
13 stations contrary to the SAE which increases or decreases suggesting changes mostly in the
14 coarse aerosol particles mode rather than in the fine mode.

15 - The analyses of the trends reported in this investigation provide evidence that both extensive
16 and intensive aerosol optical properties have significantly changed at some of the locations
17 include here over the last 10 and 15 years. The σ_{sp} decreasing trends reported here are
18 statistically significant at 5 out of 13 stations included in the analysis. These 5 stations are
19 located in the Nordic and Baltic, central and southwestern sectors. Conversely, σ_{sp} decreasing
20 trends are not statistically significant in western and eastern Europe. Statistically significant
21 decreasing trends of SAE are observed at 3 out of 10 observatories included in the analysis:
22 one site the Nordic and Baltic sector and two mountain sites in the western and eastern
23 sectors. These negative trends could be ascribed to reduced fine mode anthropogenic
24 emission as already observed in literature for columnar SAE in Europe. Conversely, at two
25 stations (one mountain site in central Europe and one urban site in southwestern Europe), the
26 SAE shows statistically significant increasing trend suggesting a shift of the accumulation mode
27 particles towards smaller sizes and/or a change in the coarse aerosol mode. At the remaining 5
28 observatories the reported SAE trends are not statistically significant. The backscatter fraction
29 shows statistically significant increasing trend at 6 out of 9 sites where BF measurements are
30 available. At three stations (the mountain site in central Europe, the urban site in southwestern
31 Europe and one of the two sites in the Nordic and Baltic sector), both BF and SAE increase
32 suggesting consistent evidence of a shift of the accumulation mode particles towards smaller
33 size. Conversely, at the other site in the Nordic and Baltic sector and at one mountain site in
34 the western sector BF increases whereas SAE decreases.

35

36 In conclusion, this investigation provides a clear and useful picture of the spatial and temporal
37 variability of the surface in-situ aerosol particle optical properties in Europe. The results presented
38 here give a comprehensive view of the particle optical properties and provide a reliable analysis of
39 aerosol optical parameters for model constraints. In addition, the analysis presented here suggests



1 findings that may need additional investigation. For example, the fact that at some of the stations
2 the trend of σ_{sp} changes in terms of both statistical significance and sign depending on the period
3 used, suggests that trend analyses are necessary in the future when longer-duration records will
4 be available. Moreover, the fact that at some sites BF and SAE show different sign in the trends
5 suggests that further analysis is needed to better understand how other aerosol parameters such
6 as particle size distribution and mean diameter affect the relationships between BF and SAE.

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1 **Tables**

2

3 **Table 1:** List of ACTRIS observatories providing aerosol particle scattering measurements

Observatory name	Country	Observatory code	Lat, Long	Altitude [m a.s.l.]	Placement (from EBAS metadata)	Inlet	Nephelometer model	Period (a)
nordic and Baltic								
Birkenes II (BIR)	Norway	NO0002R	58.3885 N, 8.252 E	219	regional	PM ₁₀	TSI3563	07/2009 –12/2015
Hyytiälä (SMR)	Finland	FI0050R	61.85N, 24.2833 E	181	regional	PM ₁₀	TSI3563	05/2006 –12/2015
Pallas (PAL)	Finland	FI0096G	67.97 N, 24.12 E	565	continental	PM ₅ ; PM _{2.5} ; PM ₁₀ (b)	TSI3563	02/2000 –12/2015
Vavihill (VHL)	Sweden	SE0011R	56.0167 N, 13.15 E	175	continental	PM ₁₀	ECOTECH Aurora3000	03/2008 –04/2014
Preila (PLA)	Lithuania	LT0015R	55.35 N, 21.0667 E	5	coastal/marine	PM ₁₀	TSI3563	12/2012 –04/2014
western								
Mace Head (MHD)	Ireland	IE0031R	53.3258 N, -9.8994 E	5	coastal/marine	whole air	TSI3563	07/2001 –12/2013
Cabauw (CBW)	The Netherlands	NL0011R	51.9703 N, 4.9264 E	1	regional	PM ₁₀	TSI3563	01/2008 –12/2012
SIRTA (SIR)	France	FR0020R	48.7086 N, 2.1589 E	162	suburban	whole air	ECOTECH M9003	07/2012 –12/2013
Observatory Perenne (OPE)	France	FR0022R	48.5622 N, 5.505555 E	392	regional	whole air; PM ₁₀ (c)	ECOTECH Aurora3000	09/2012 –12/2015
Puy de Dome (PUY)	France	FR0030R	45.7667 N, 2.95 E	1465	mountain	whole air	TSI3563	01/2007 –12/2014
central								
Hohenpeissenberg (HPB)	Germany	DE0043G	47.8 N, 11.0167 E	985	mountain	PM ₁₀	TSI3563	01/2006 –12/2015
Ispra (IPR)	Italy	IT0004R	45.8 N, 8.6333 E	209	semi regional	PM ₁₀	TSI3563	01/2004 –12/2014
Melpitz (MPZ)	Germany	DE0044R	51.53 N, 12.93 E	86	regional	PM ₁₀	TSI3563	01/2007 –12/2015
Jungfraujoch (JFJ)	Switzerland	CH0001G	46.5475 N, 7.985 E	3578	mountain	whole air	TSI3563	07/1995 –12/2015
Mt. Cimone (CMN)	Italy	IT0009R	44.1833 N, 10.7 E	2165	mountain	whole air	ECOTECH Aurora M9003; TSI 3563 (d)	05/2007 –12/2015
Koštice (KOS)	Czech Republic	CZ0007R	49.58333N, 15.0833 E	534	regional	PM ₁₀	TSI3563	03/2013 – 12/2015
eastern								
Beo Moussala (BEO)	Bulgaria	BG0001R	42.1667 N, 23.5833 E	2971	mountain	whole air	TSI3563	03/2007 –12/2015
K-Puszta (KPS)	Hungary	HU0002R	46.9667 N, 19.5833 E	125	regional	PM ₁ ; PM ₁₀ (e)	TSI3563	05/2006 –12/2014
south-western								
Montsec (MSA)	Spain	ES0022R	42.0513 N, 0.44 E	1570	mountain	PM _{2.5} ; PM ₁₀ (f)	ECOTECH Aurora3000	01/2013 – 12/2015
Izaña (IZO)	Spain	ES0018G	28.309 N, -16.4994 E	2373	mountain	PM ₁₀	TSI3563	03/2008 – 12/2015
Granada (UGR)	Spain	ES0020U	37.164 N, -3.605 E	680	urban	whole air	TSI3563	01/2006 –12/2015
Montseny (MSY)	Spain	ES1778R	41.7667 N, 2.35 E	700	regional	PM ₁₀	ECOTECH Aurora3000	01/2010 –12/2015
Madrid (MAD)	Spain	ES1778R	40.4627 N, -3.717 E	669	sub-urban	PM _{2.5} ; PM ₁₀ (g)	ECOTECH Aurora3000	01/2014 – 12/2014
south-eastern								
Finokalia (FKL)	Greece	GR0002R	35.3167 N, 25.6667 E	250	coastal/marine	whole air; PM ₁ ; PM ₁₀ (h)	RR M903; Ecotech Aurora1000 (i)	04/2004 –12/2015
Athens (DEM)	Greece	GR0100B	37.9905 N, 23.8095 E	270	sub-urban	PM ₁₀	ECOTECH Aurora3000	01/2012 –12/2015
Arctic								



Zeppelin (ZEP)	Svalbard (Norway)	NO0042G	78.9067 N, 11.8883 E	474	arctic environment	PM ₁₀	TSI3563	07/2010 – 12/2014
Antarctic								
Troll (TRL)	Antarctica	NO0058G	-72.0167 N, 2.5333 E	1309	antarctic environment	whole air; PM ₁₀ (j)	TSI3563	02/2007 – 12/2015
South America								
Mt. Chacaltaya (CHC)	Bolivia	BO0001R	-16.2000 N, -68.09999 E	5240	mountain	whole air	ECOTECH Aurora3000	01/2012 – 12/2015 (k)

1 (a) Start-end of measurements; Total aerosol particle scattering was used as reference for measurement period; (b) PM₅ (2000-2005),
2 PM_{2.5} (2005-2008) and PM₁₀ (2008-2015); (c) whole air (2012-2013) and PM₁₀ (2014-2015); (d) ECOTECH Aurora M9003 during 2007-
3 2013 and TSI 3563 (2014-2015); (e) PM₁ (2006-04/2008) and PM₁₀ (05/2008-2014); (f) PM_{2.5} (2013) and PM₁₀ (2014-2015); (g) PM₁₀ from
4 03/2014; (h) whole air (2004-2008), PM₁₀ (2009-2011), PM₁ (2011-2012), PM₁₀ (2013-2015); (i) RR M903 during 2004-2011, Ecotech
5 AURORA1000 during 2012-2015; (j) whole air (2007-2009) and PM₁₀ (2010-2015); (k) Only measurements performed during the year
6 2012 were used in this investigation.
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1 **Table 2:** Trends of aerosol particle scattering coefficient (σ_{sp}), scattering Ångström exponent (SAE),
 2 and backscatter fraction (BF). Three trends for SAE are reported: SAE calculated as linear fit using
 3 three wavelengths (b-g-r); using the blue and the green wavelengths (b-g) and using the green and
 4 red wavelengths (g-r). Trend results are reported for the whole period available at each station until
 5 2015 (bold) and for the periods reported in Collaud Coen et al. (2013) and in Asmi et al. (2013).
 6 Trends are considered as statistically significant if p-value < 0.05. Statistically significant increasing
 7 or decreasing trends are highlighted with up (↑) and down (↓) red and green arrows, respectively.
 8 Non-statistically significant increasing or decreasing trends are highlighted with up (↗) and down (↘)
 9 grey arrows, respectively. Grey colored table cells highlight stations included in this work but not
 10 included in the works from Collaud Coen et al. (2013) or Asmi et al. (2013). \$: parameters removed
 11 in this work and in the work from Collaud Coen et al. (2013) because of measurement gaps, low
 12 data coverage or break points for one or more wavelengths. #: Only available for 2014-2015; ± not
 13 available.

Station	period	Trend (This work)				MK Trend (Collaud Coen et al., 2013)				MK Trend (Asmi et al., 2013)				
		σ_{sp}	SAE			BF	σ_{sp}	SAE			BF	Particle number		
			b-g-r	b-g	g-r			b-r	b-g	g-r		N	N20 (20-500 nm)	N100 (100-500 nm)
Nordic and Baltic														
PAL	2000 - 2015	↗	↘	↘	↘	↗								
	2000 - 2010	↘	↘	\$	\$	↗	↘	↗	\$	\$	↗			
	2001 - 2010	↘	↘	\$	\$	↗	↘	↗	\$	\$	↗	↘ (10-500 nm)	↔	
	1996 - 2010											↘ (10-500 nm)		
SMR	2006 - 2015	↘	↗	↗	↗	↗								
	1996 - 2011											↘	↘	
	2001 - 2010											↘	↘	
western														
MHD	2001 - 2013	↘	\$	\$	\$	\$								
	2000 - 2010											↘ (3-500 nm)		
	2001 - 2010	↗	\$	\$	\$	\$	↗	\$	\$	\$	↗	↗ (3-500 nm)		
PUY	2007 - 2014	↘	↘	↘	↘	↗								
central														
HPB	2006 - 2015	↘	↗	↗	↗	↗								
	2001 - 2010						↗	\$	\$	\$	\$			
	2002 - 2010						↘	\$	\$	\$	\$			
	1995 - 2011											↗ (15-500 nm)		
IPR	2004 - 2014	↘	↗	↗	↗	↗								
MPZ	2007 - 2015	↘	↘	↘	↘	↗								
	1997 - 1998 and 2004 - 2010											↗	↗	
JFJ	1995 - 2015	↘	\$	\$	\$	\$								
	1995 - 2010	↗	\$	\$	\$	\$	↗	\$	\$	\$	\$			
	1996 - 2010	↗	\$	\$	\$	\$	↗	\$	\$	\$	\$			
	2001 - 2010	↘	\$	\$	\$	\$	↘	\$	\$	\$	\$	↘ (10-500 nm)		
	1997 - 2010	↗	\$	\$	\$	\$						↗ (10-500 nm)		
CMN	2007 - 2015	↘	#	#	#	#								
eastern														
BEO	2007 - 2015	↘	↘	↘	↘	↘								



KPS	2006 - 2014	↑	↓	↓	↑	↑								
<i>south-western</i>														
IZO	2008 - 2015	↓	↑	↑	↑	\$								
UGR	2006 - 2015	↓	↑	↑	↑	↑								

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5 **Table 3:** Daytime (08:00 – 16:00 GMT) and nighttime (21:00 – 05:00 GMT) of σ_{sp} trends by season calculated
 6 for the periods considered in this work. Sp: Spring; Su: Summer; Au: Autumn; Wi: Winter. Trends are
 7 considered as statistically significant if p-value < 0.05. Statistically significant increasing or decreasing trends
 8 are highlighted with up (↑) and down (↓) red and green arrows, respectively. Non statistically significant
 9 increasing or decreasing trends are highlighted with up (↑) and down (↓) grey arrows, respectively.

Station	period	SCATTERING					
		daytime		nighttime		24h	
		Sp	Su	Sp	Su	Sp	Su
		Au	Wi	Au	Wi	Au	Wi
JFJ	1995 - 2015	↓	↓	↓	↓	↓	↓
		↑	↓	↑	↓	↑	↓
HPB	2006 - 2015	↓	↓	↓	↓	↓	↓
		↓	↓	↓	↓	↓	↓
PUY	2006 - 2014	↓	↓	↓	↓	↓	↓
		↓	↓	↓	↓	↓	↓
CMN	2007 - 2015	↓	↑	↓	↓	↓	↓
		↓	↓	↓	↓	↓	↓
BEO	2007 - 2015	↓	↓	↓	↑	↓	↓
		↓	↓	↓	↑	↓	↑
IZO	2008 - 2015	↓	↓	↓	↓	↓	↓
		↑	↓	↑	↓	↑	↓

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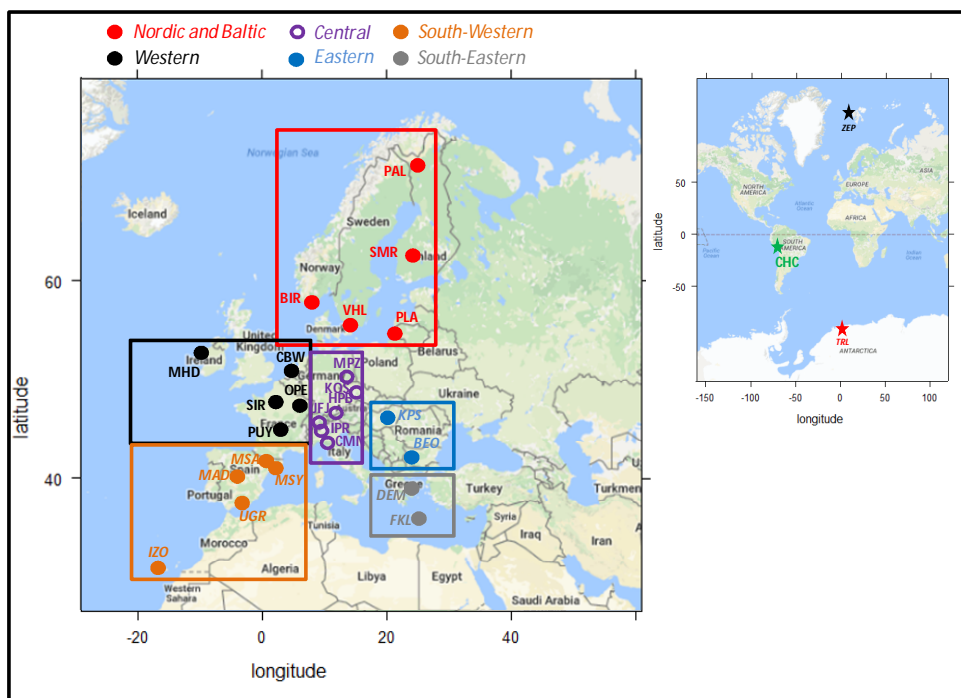
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1 **Figures**

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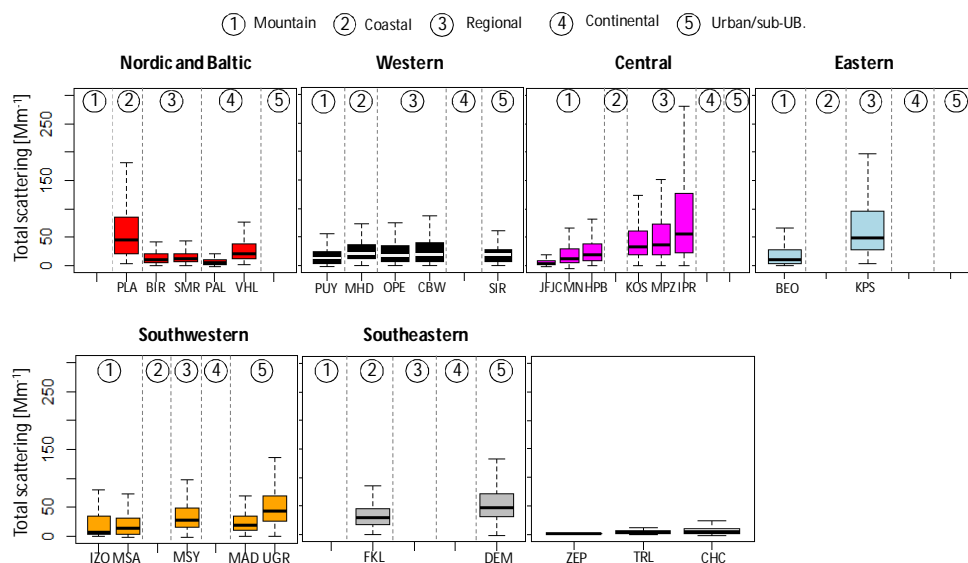
6 **Figure 1:** Location of the 28 ACTRIS stations included in this work.

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Figure 2: Total aerosol scattering coefficients in the green divided by geographical location. At SIR aerosol scattering was available only at 450 nm. Medians (horizontal lines in the boxes), percentiles 25th and 75th (lower and upper limits of the boxes, respectively) and percentiles 5th and 95th (lower and upper limits of the vertical dashed lines) are reported. Hourly data were used for the statistic. For each location data are ordered from mountain sites (1) to urban/sub-urban sites (5).

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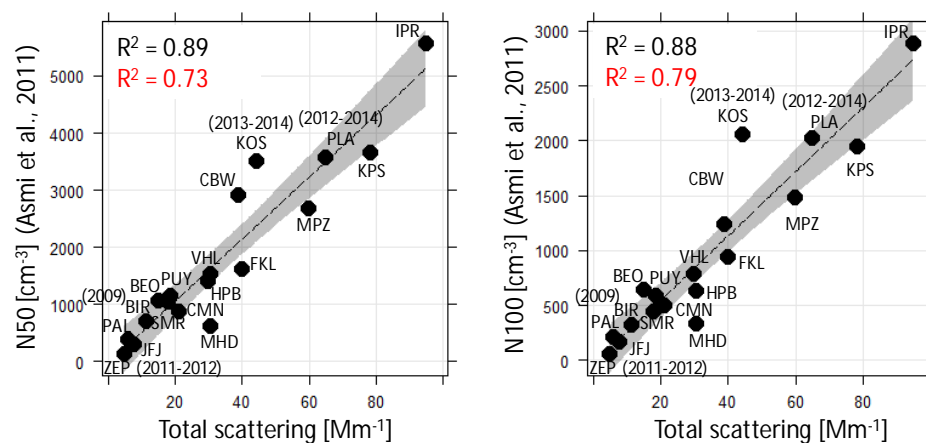
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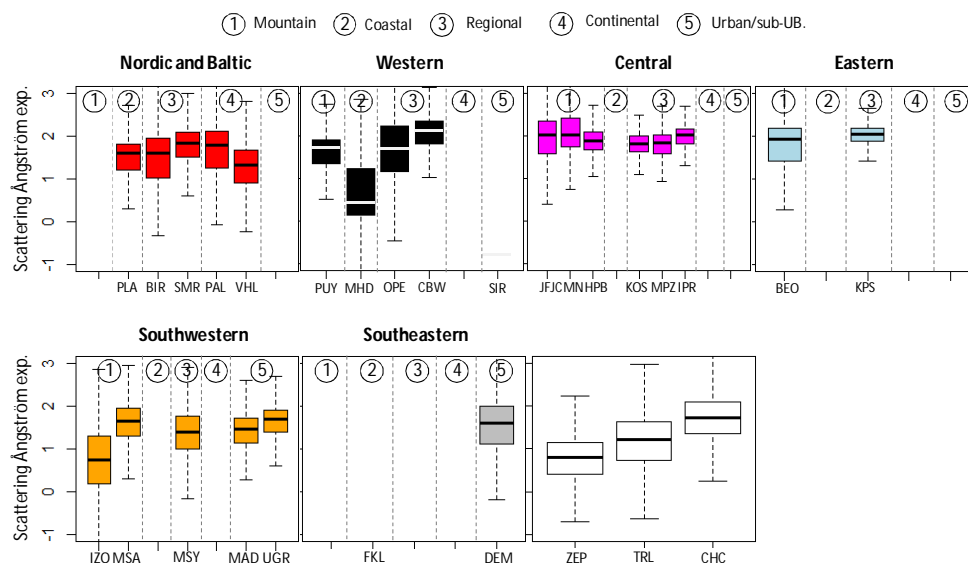
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Figure 3: Relationship between N50 (mean particle number concentration between 50 nm and 500 nm) and N100 (mean particle number concentration between 100 nm and 500 nm) and mean aerosol particle scattering coefficient averaged over the period 2008 – 2009. For ZEP, BIR, KOS and PLA aerosol particle scattering measurements were not available during 2008 – 2009 and different period were used. R² highlighted in red were obtained using the median values.

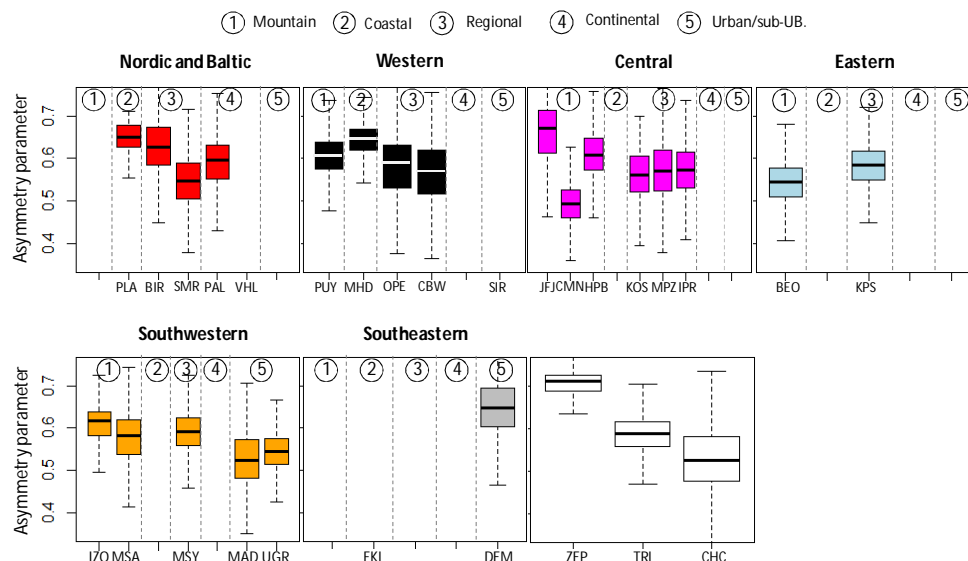
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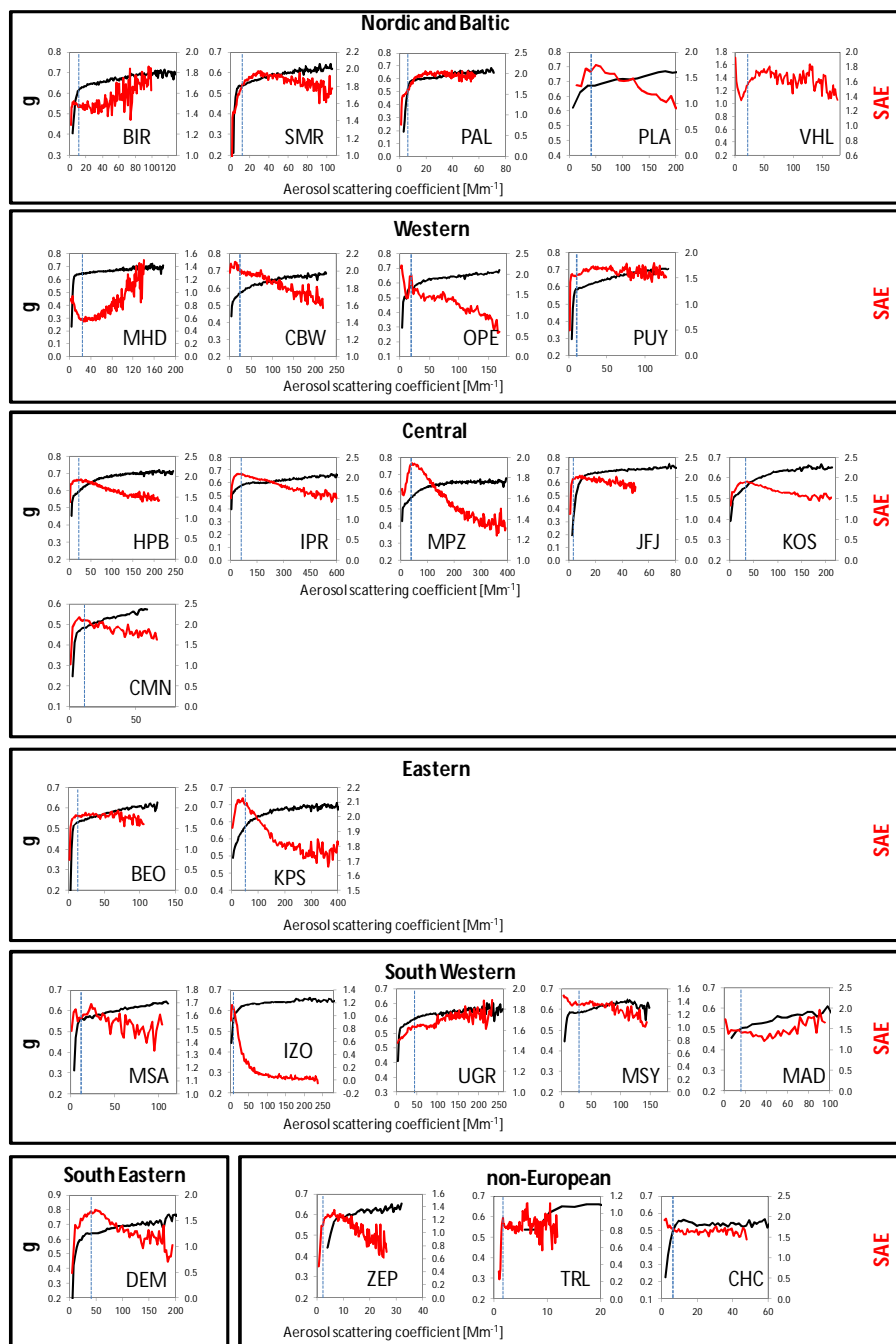
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Figure 4: Scattering Ångström exponent divided by geographical location. Medians (horizontal lines in the boxes), percentiles 25th and 75th (lower and upper limits of the boxes, respectively) and percentiles 5th and 95th (lower and upper limits of the vertical dashed lines) are reported. For each location data are ordered from mountain sites to urban/sub-urban sites. At CHC, the SAE was calculated using the blue and green wavelengths.



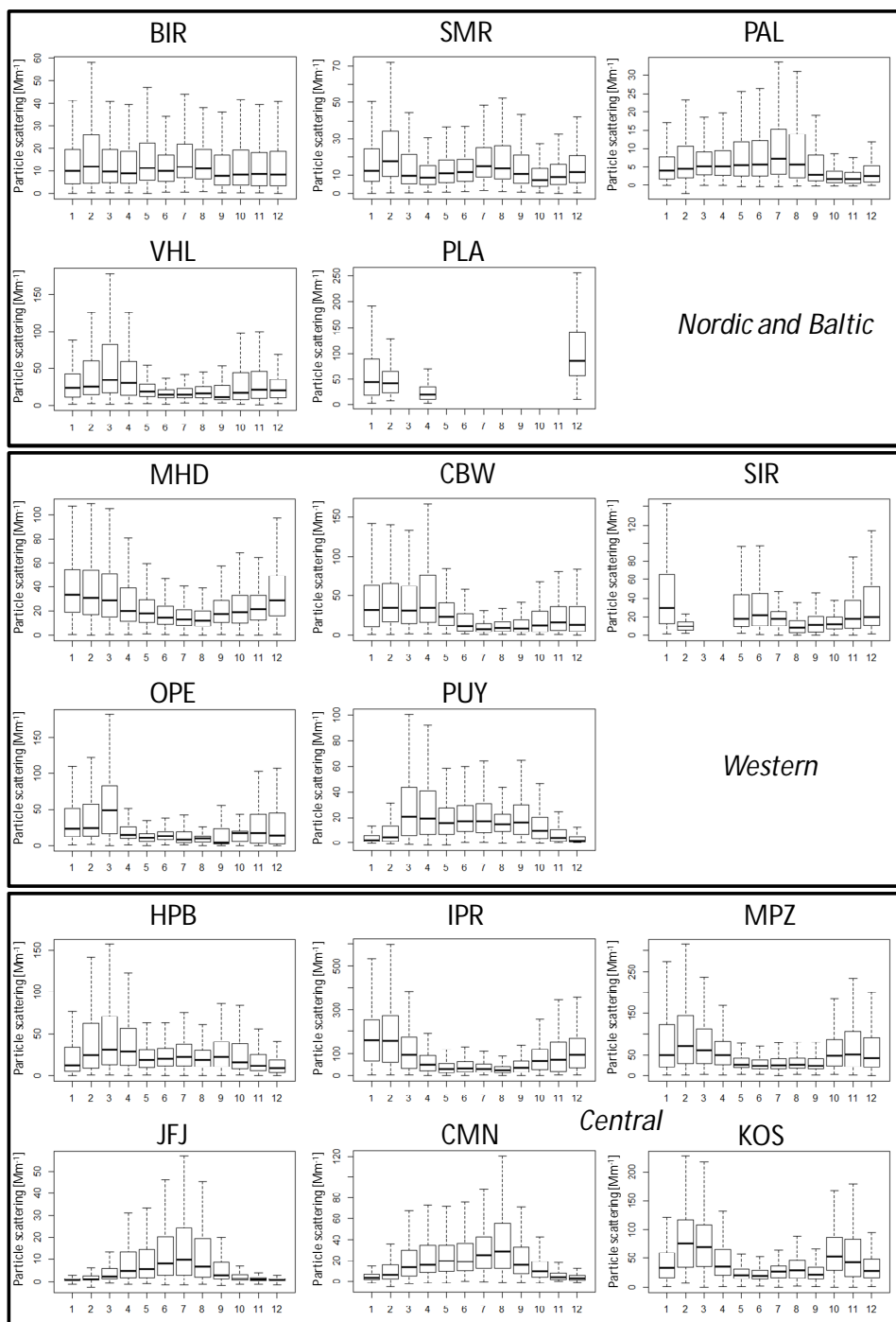
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Figure 5: Asymmetry parameter in the green divided by geographical location. Medians (horizontal lines in the boxes), percentiles 25th and 75th (lower and upper limits of the boxes, respectively) and percentiles 5th and 95th (lower and upper limits of the vertical dashed lines) are reported. For each location data are ordered from mountain sites to urban/sub-urban sites.



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Figure 6: Scatterplots between σ_{sp} (x-axes) and SAE (right y-axes; red lines) and g (left y-axes; black lines). Dashed lines represent median σ_{sp} values at each station. At CHC, SAE was calculated using the blue and the green wavelengths.



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Figure 7: Seasonal cycles of σ_{sp} [Mm^{-1}] measured in the green nephelometer wavelength.

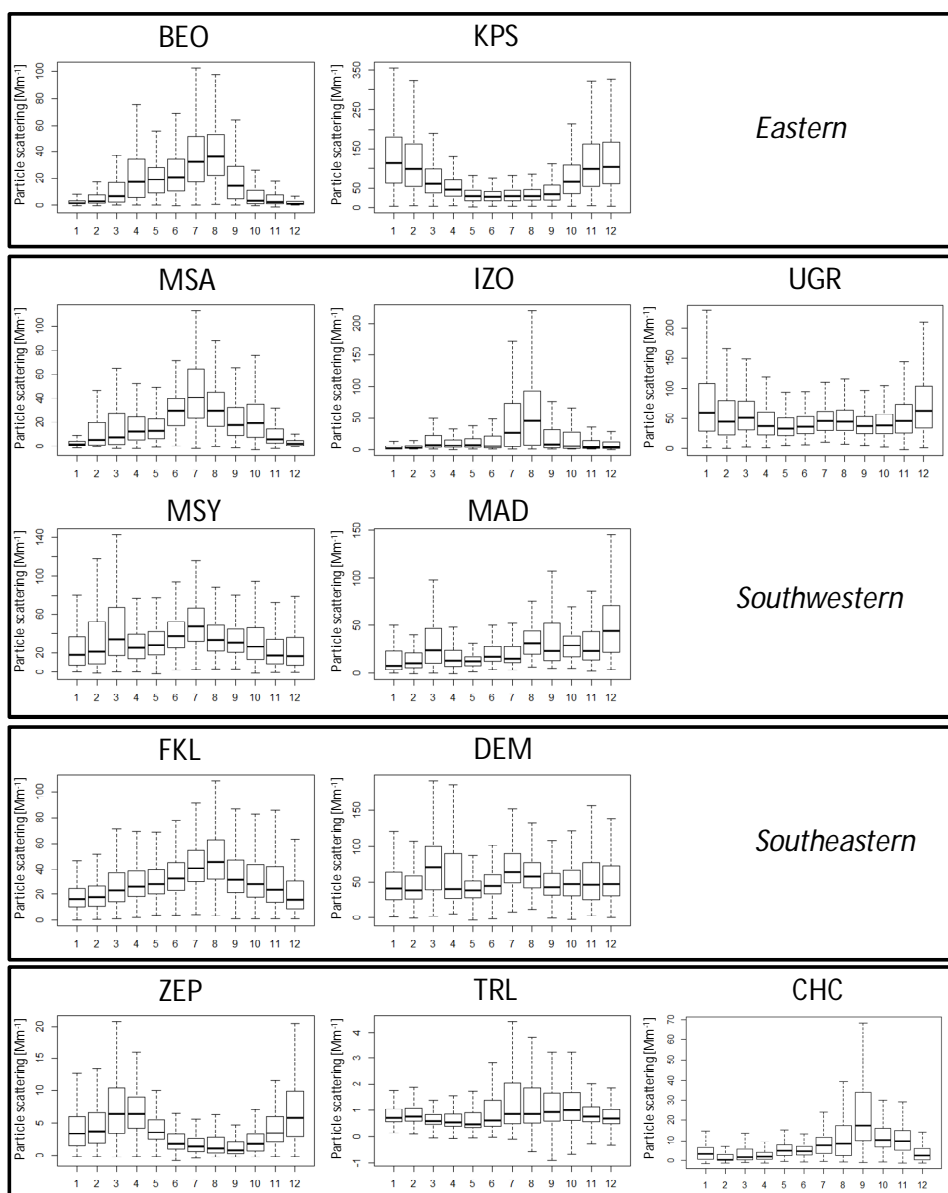
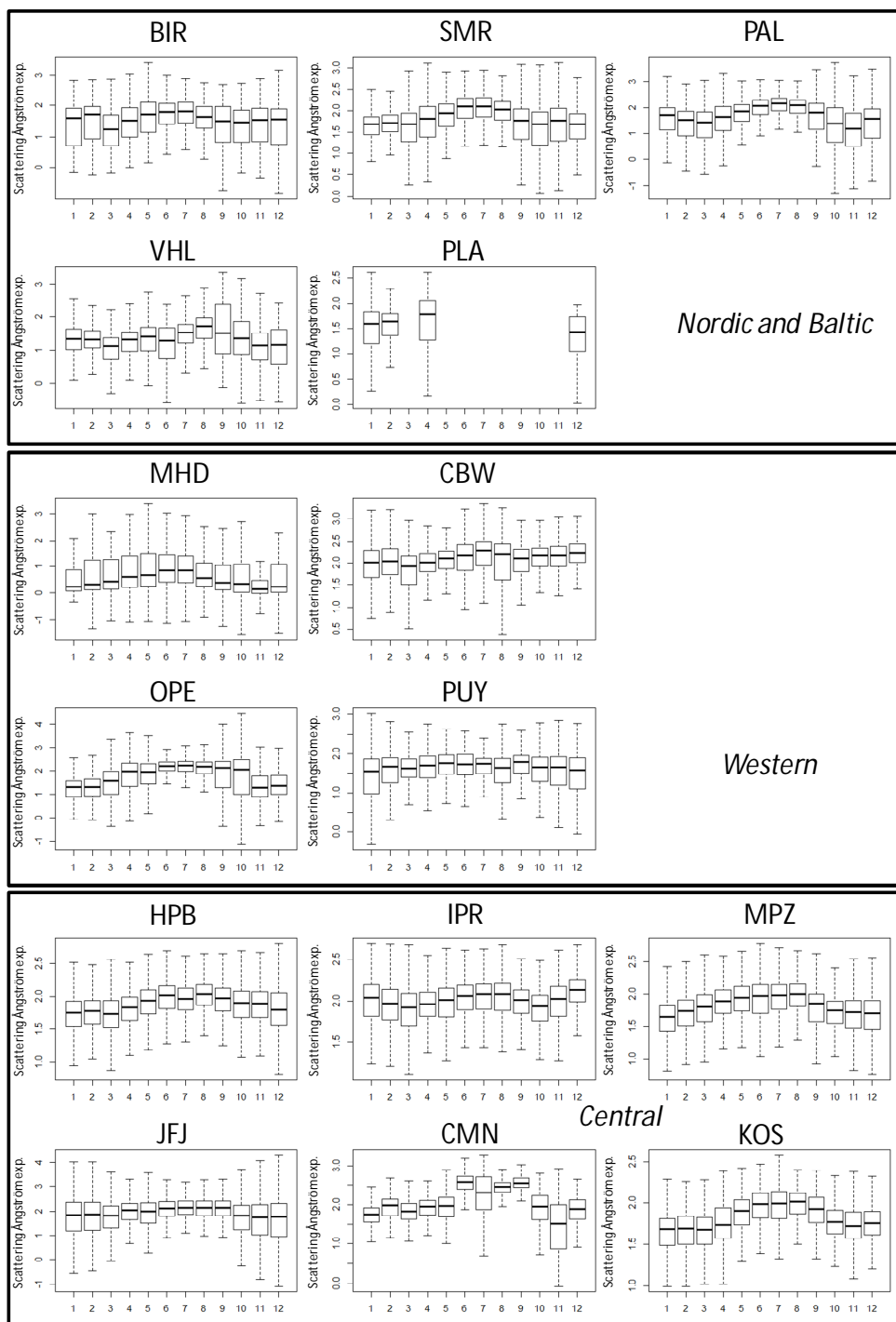


Figure 7: (Continued) Seasonal cycles of σ_{sp} [Mm^{-1}] measured in the green nephelometer wavelength.

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Figure 8: Seasonal cycles of SAE (calculated as linear fit using three nephelometer wavelengths)

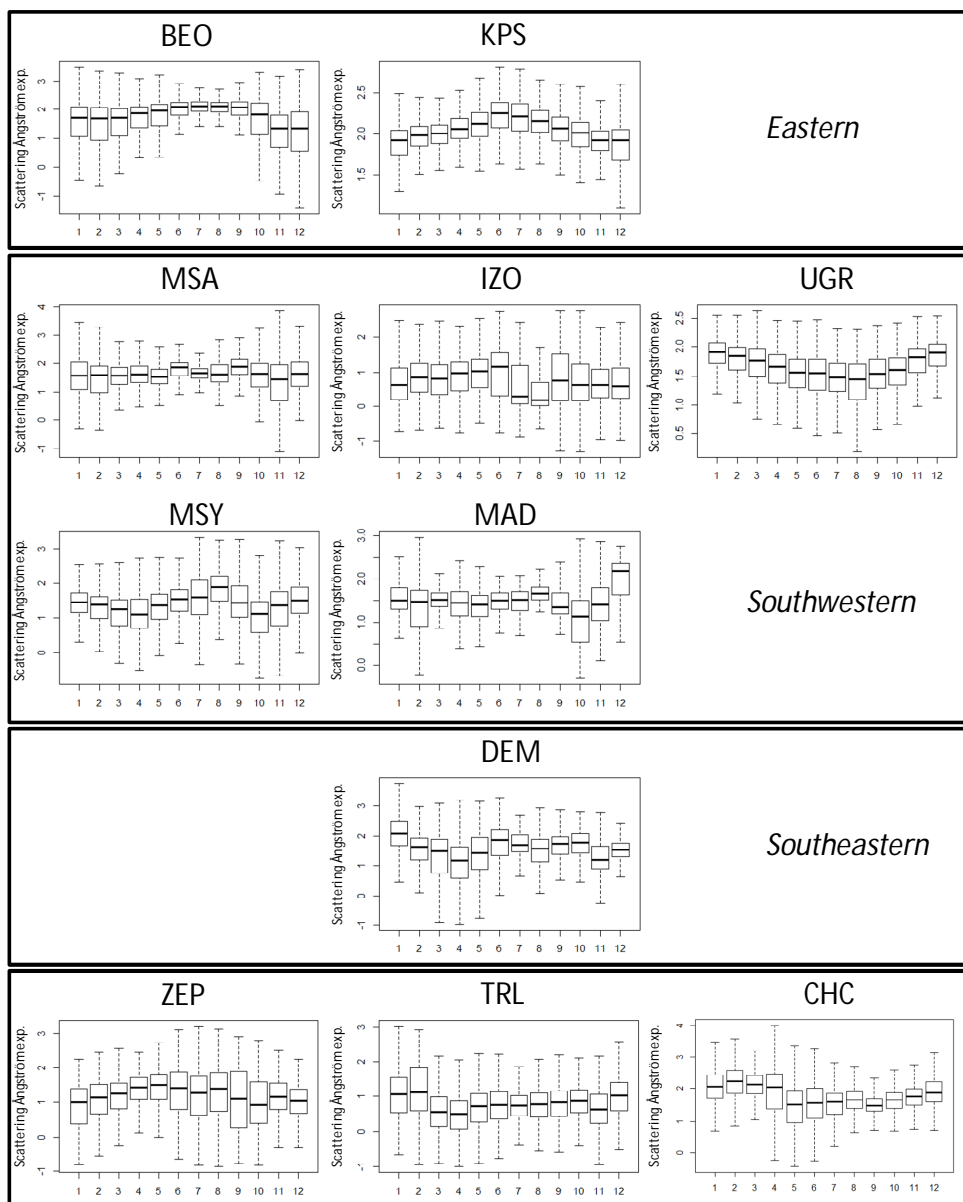
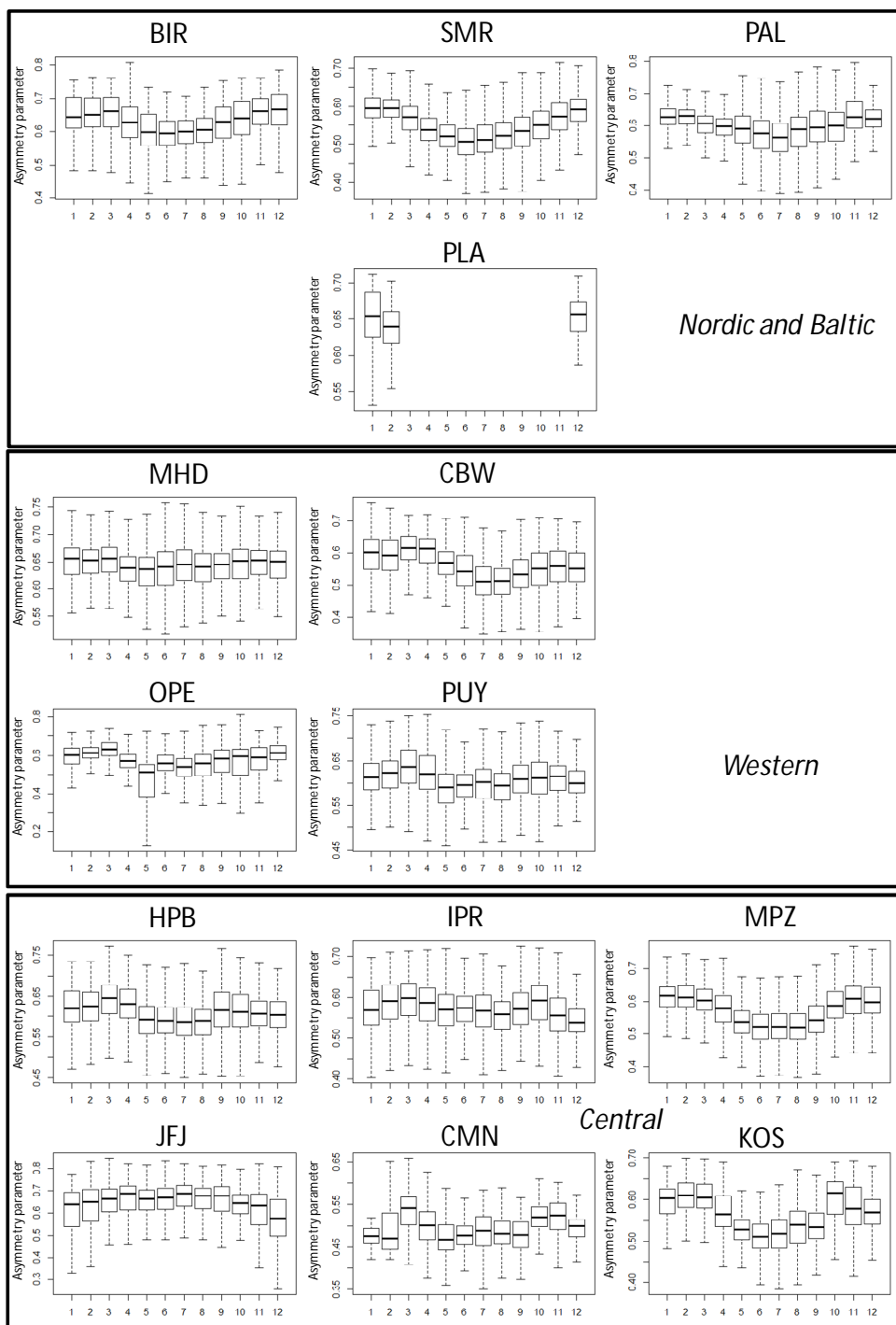


Figure 8: (Continued) Seasonal cycles of SAE (calculated as linear fit using three nephelometer wavelengths). At CHC the SAE was calculated using the blue and the green wavelengths.

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Figure 9: Seasonal cycles of g (calculated for the green wavelength).

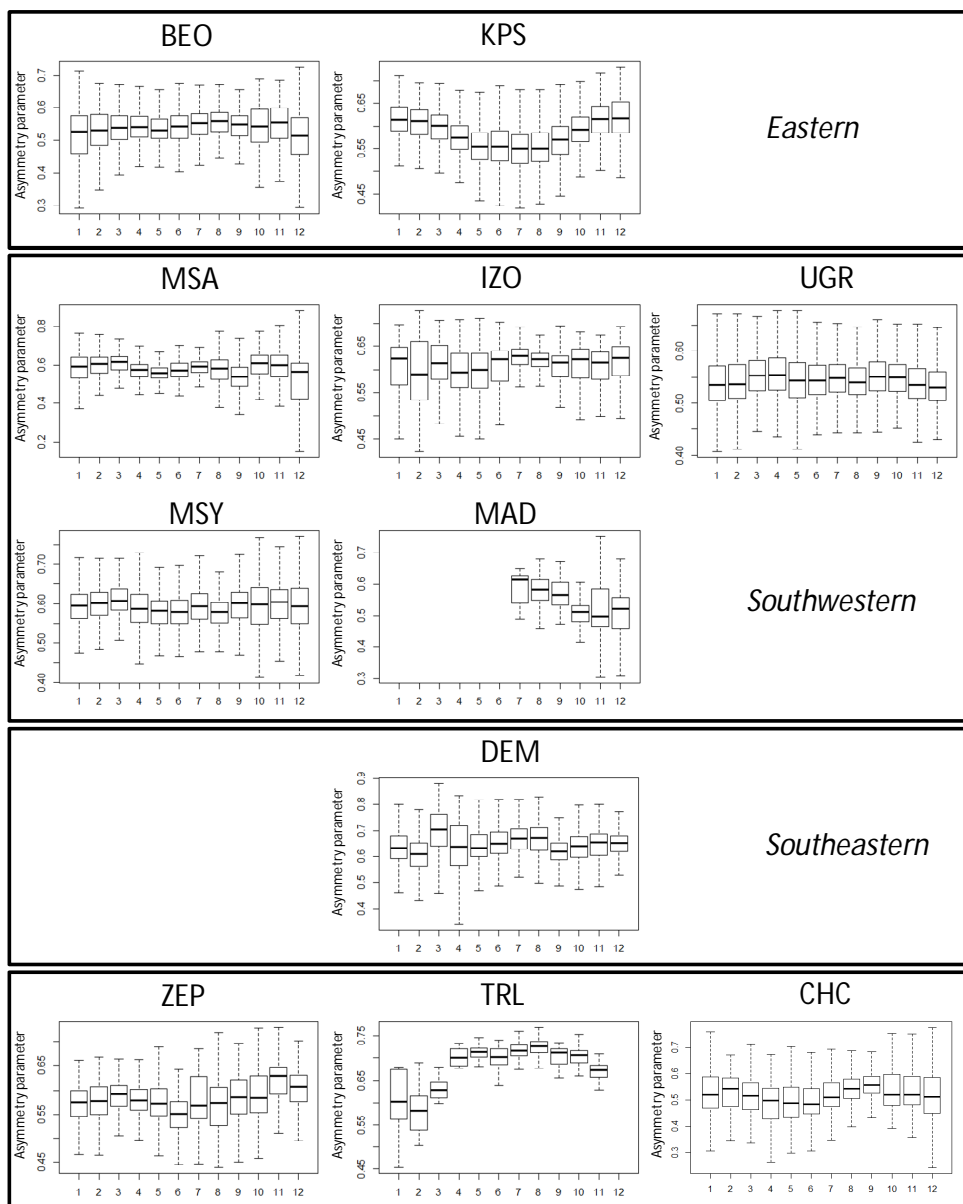


Figure 9: (Continued) Seasonal cycles of g (calculated for the green wavelength).

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