

Dear editor, dear Referees, We would like to thank you for all your comments. This input has allowed us to refine the manuscript by adding more thorough detailed explanations, to correct some points and to improve in a large sense the manuscript.

Below the answers to the Referee's comments

Review of Pandolfi et al 2017 – ACTRIS nephelometer measurements. This manuscript summarizes nephelometer measurements (in terms of monthly climatology, covariance of scattering-related optical properties and trends) across the ACTRIS network in Europe. The manuscript also includes a few non European sites supported by ACTRIS. A lot of work went into bringing these data sets together and summarizing station differences (instrumentation, size cut, corrections...).

General/Technical comments

The manuscript includes a lot of information and clearly a lot of work was involved in getting that information into useable form for analysis, but, as written, it's really hard to digest. Some of that would be helped by more editing (e.g., shorter paragraphs as described in my editorial comments below).

1) Why not compare scattering seasonality with Zanatta seasonality of absorption where possible?

Figure 3 was modified to include a comparison of total scattering coefficients with the absorption coefficients reported in Zanatta et al. (2016; Table 8). The text describing Figure 3 has been accordingly modified.

“The observed variation is consistent with the differences in particulate matter (PM) mass concentrations, PM chemical composition, particle number concentration and absorption coefficients observed across Europe, as described for example by Putaud et al. (2010), Asmi et al. (2011) and Zanatta et al. (2016).

Figures 3a and 3b show the relationship between the mean particle number concentration measured at different stations during 2008 to 2009 (and reported in Asmi et al. (2011)) and the mean σ_{sp} measured over the same period (where available). As reported in Figure 3, good correlations are observed between N50 (Figure 3a: mean/median particle number between 50 nm and 500 nm) and N100 (Figure 3b: mean/median particle number between 100 nm and 500 nm) and mean σ_{sp} . Figure 3c shows the relationship (for some stations) between absorption coefficients reported in Zanatta et al. (2016) and the total scattering. The good correlations reported in Figure 3c (especially high for the winter and autumn periods) suggest an increase of both scattering and absorption coefficients with increasing aerosol loading.”

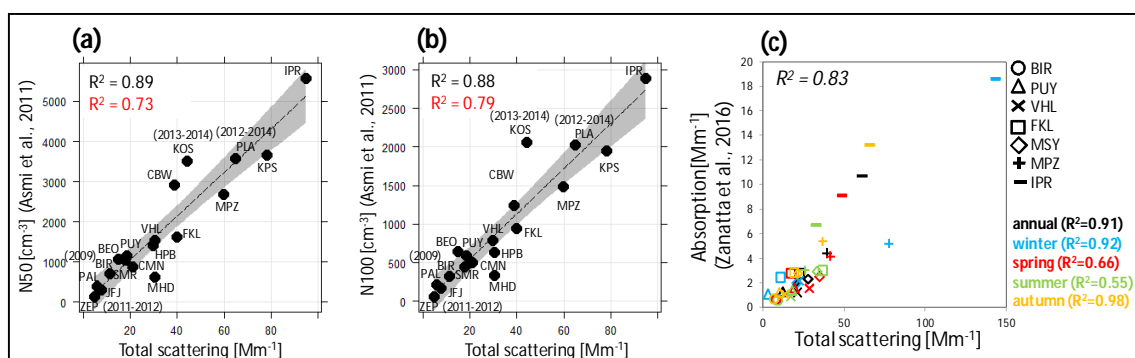


Figure 3: Relationship between: (a) N50 (mean particle number concentration between 50 nm and 500 nm), (b) N100 (mean particle number concentration between 100 nm and 500 nm), (c) absorption coefficient and mean aerosol particle total scattering coefficient. (a) and (b): data averaged over the period 2008 to 2009. For ZEP, BIR, KOS and PLA aerosol particle scattering measurements were not available during 2008 to 2009 and different periods were used. R2 values, highlighted in red, were obtained using the median values. (c) Data averaged as in Zanatta et al. (2016).

- 2) ***Why not present SSA seasonality/trends at the same time? I realize an absorption measurement is needed to do so – but Zanatta’s ACTRIS paper means those data exist for some sites and should be in some sort of consistent form.***

Certainly, absorption coefficient measurements are available at the majority of the observatories included in the present manuscript. However, a manuscript presenting SSA climatology and trends at ACTRIS sites is already under preparation. Moreover, we think that adding SSA (and consequently absorption) would make the manuscript too long and difficult to read. For this reason and with the permission of the Reviewer we prefer not to include SSA in the present manuscript.

- 3) ***I understand you want to include all ACTRIS supported light scattering measurements, but the non-European sites (TRL and CHC) are a distraction. They seem to only be included because they are part of ACTRIS but not for any strong scientific reason and are barely discussed in the text. I think it is fine to include these stations in the supplemental materials if you must, but they should be eliminated from the main manuscript.***

Following the Reviewer suggestion, the figures related to the two non-European sites TRL and CHC were moved to the supporting material. The Figure is reported below:

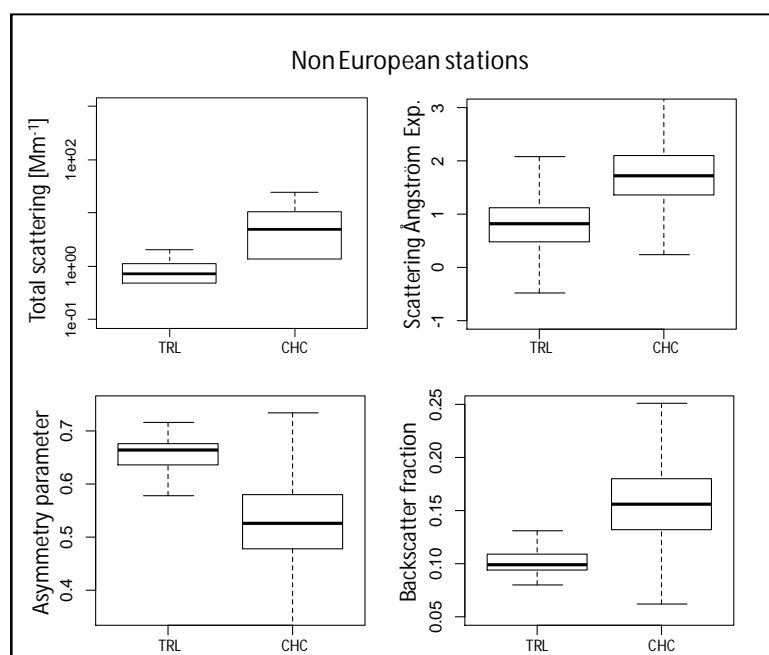


Figure S3: Total scattering, Scattering Angstrom Exponent, Asymmetry parameter and Backscatter fraction for the non-European stations TRL and CHC.

- 4) ***Page 6, Line 23 – the nomenclature ‘regional’ and ‘continental’ is a bit confusing. Perhaps change continental to remote or rural? Or combine to ‘continental’ and note that some are more polluted than others? This would make sense as I think they are usually discussed together in the manuscript.***

We thank the Reviewer for this comment which considerably simplifies the discussion. As suggested by the Reviewer the PAL station was included in the “Arctic” category in the revised version of the

manuscript. Thus, following the EBAS definition, the only continental station is VHL. Consequently, the category "continental" was removed in the revised version of the manuscript and VHL was included in the new "regional/rural" category.

- 5) Page 7 – somewhere in Section 2 (under data treatment) you should explain where the data came from, i.e., was it downloaded directly from EBAS (if so, what level (presumably level 2)). Or was it provided by data providers? Or some combination of the two? You should also say whether you performed additional quality checks on the data or did you take them "as is" from EBAS/data providers? I would hesitate to assume that even Level 2 data downloaded from EBAS it is OK to use without further review (particularly if you are working with backscatter values of any wavelength and blue or red scattering values. Personal experience with the Level 2 nephelometer data in EBAS suggests that much of the data provider QC focuses primarily on the green scattering and the other nephelometer parameters may not get as much attention. A further issue is that it seems that often data QC is done by different individuals in different years and they don't look at years before and after their year to see if there are obvious differences.**

In order to take into account the Reviewer comments #5 and #53, the section 2 was modified. Thus, the following sentences were added at the end of the section 2.2.1 and beginning of section 2.2.2:

"Recommended quality assurance procedures during on-site operation, as described in GAW (WMO-GAW Report, 2016), help to ensure the quality and comparability of the data. The nephelometers included in this investigation are regularly calibrated using span gas and are zero adjusted using particle-free air. Additionally, most of the integrating nephelometers employed in ACTRIS have undergone a schedule of performance checks at the World Calibration Center for Aerosol Physics of ACTRIS/GAW."

"2.2.2 Data treatment

Data used in this investigation include hourly averaged Level 2 aerosol particle scattering data downloaded from the ACTRIS/EBAS Data Centre web portals (www.actris.nilu.no; www.ebas.nilu.no; last downloads August 2017). The σ_{sp} and σ_{bsp} data reported to EBAS and used in this work are referenced to standard T (273.15 °C) and P (1013 hPa) conditions. Data consistency is critical when comparing many years' worth of data from different stations. In this work, the Level 2 scattering data were further reviewed in order to ensure a high quality of the data presented. There are however station-to-station differences (e.g. sizecut, RH control, wavelength, data processing, etc.) which are addressed in the sections below."

Moreover, during the discussion further feedback was needed with data providers from some stations regarding the correction of scattering data. Now, all scattering data presented in the revised version of the manuscript are corrected for truncation with the exception of FKL, CMN, and SIR (where only $1-\lambda$ was available; see also the authors reply to the Reviewer comment #9). The section 2.2.2.1 was accordingly modified as follow:

"2.2.2.1 Truncation correction

Data from the integrating nephelometers used here are corrected for non-ideal illumination of the light source (deviation from a Lambertian distribution of light) and for truncation of the sensing volumes in the near-forward (around 0-10°) and near-backward (around 170-180°) directions (Müller et al., 2009 and Anderson and Ogren, 1998). Correction schemes have been provided by Müller et al. (2009; 2011) for the RR M903 and Ecotech models M9003 and AURORA3000, and by Anderson and Ogren (1998) for the TSI3563. These schemes consist of a simple linear correction based on the scattering Ångström exponent (SAE) determined from the raw nephelometer data to take account of the size-distribution-dependent truncation error. It has been demonstrated that these simple correction schemes are accurate for a wide range of atmospheric aerosols and that the uncertainties in the corrections are not expected to be larger than 2% for an aerosol particle population with a single scattering albedos (SSA) greater than 0.8 (Bond et al., 2009).

The majority of the σ_{sp} data in the EBAS database are corrected for non-ideal illumination and for truncation by the data providers. Exceptions are the scattering data submitted for KOS, MHD, PLA, CMN, FKL and SIR. Scattering data from KOS, MHD and PLA were corrected in this work using the correction scheme provided by Anderson and Ogren (1998) (cf. Table S1 of the Supporting Material). The σ_{sp} data collected at CMN, FKL and

SIR are not corrected because the nephelometers deployed at these three stations provide scattering only at one wavelength, thus preventing the estimation of the SAE. Given that the nephelometer correction factors vary as a function of SAE, the assumption of a constant correction factor to correct the $1-\lambda$ scattering data could introduce undesired noise. Moreover, at SIR and CMN, the σ_{sp} is measured with the single wavelength Ecotech nephelometer model M9003 (until 2013 at CMN). The correction curve from Müller et al. (2009; Figure 4) provides a correction factor of around 0.97 to 1.0 for the M9003 for a SAE of around 1.5 to 2. Using the TSI3563 scattering measurements performed at CMN during 2014-2015, we estimated a mean SAE of around 2 for CMN (cf. Table S5). Thus, given the rather small effect of the correction factor estimated for the Ecotech M9003, scattering data from CMN and SIR were not corrected in this work. At FKL the nephelometer models RR M903 (until 2011) and Ecotech 1000 (from 2012) were used (cf. Table 1). To the best of our knowledge, no correction scheme has been provided for the Ecotech 1000. Moreover, at FKL, the inlet was changed many times (cf. Table 1) and the correction factors provided in the literature are a strong function of the size cut-off used. For these reasons, scattering data collected at FKL are not corrected in this investigation."

- 6) Page 7, Line 15-18 – Later on you note that there are differences in operations from the GAW protocols – the measurement RH is one obvious difference from GAW protocols at many sites, so the statement ‘the nephelometer instruments are run following the ACTRIS/GAW standards (WMO-GAW Report, 2016)’ is not strictly true. Please revise.**

In the revised version of the manuscript we removed the sentence at Pag. 7, Line 15-18:

"The stations included in this work report the data to ACTRIS and GAW/EMEP, consequently the data are quality assured given that the nephelometer instruments are run following the ACTRIS/GAW standards (WMO-GAW Report, 2016) and regularly inter-compared."

We removed the sentence because it is a repetition: The sentence is moved at the end of the Paragraph 2.2.1 and modified as follow (see also the replies to the Reviewer comments #5 and #54):

"Recommended quality assurance procedures during on-site operation, as described in GAW (WMO-GAW Report, 2016), help to ensure the quality and comparability of the data. The nephelometers included in this investigation are regularly calibrated using span gas and are zero adjusted using particle-free air. Additionally, most of the integrating nephelometers employed in ACTRIS have undergone a schedule of performance checks at the World Calibration Center for Aerosol Physics of ACTRIS/GAW."

"2.2.2 Data treatment

Data used in this investigation include hourly averaged Level 2 aerosol particle scattering data downloaded from the ACTRIS/EBAS Data Centre web portals (www.actris.nilu.no; www.ebas.nilu.no; last downloads August 2017). The σ_{sp} and σ_{bsp} data reported to EBAS and used in this work are referenced to standard T (273.15 °C) and P (1013 hPa) conditions. Data consistency is critical when comparing many years' worth of data from different stations. In this work, the Level 2 scattering data were further reviewed in order to ensure a high quality of the data presented. There are however station-to-station differences (e.g. sizecut, RH control, wavelength, data processing, etc.) which are addressed in the sections below."

- 7) Page 7, Line 29 – ‘Due to the non-homogeneity of the light source’ this should be made clearer. Was the light source causing measurement problems and is there a reference report describing this? Do you use the data from before the light source change? Why were different replacement light sources used at SIR and CMN and does this have implications for the data?**

The problems with the angular intensity function of the nephelometer Ecotech model M9003 have been discussed by Muller et al. (2009). This reference was added to the sentence at Pag. 7, Line 29. Yes, in the manuscript we also used the scattering data collected before the change of the light source. We do not know why different replacements were used for SIR and CMN, but both data providers confirmed (personal communication) that both nephelometers performed well after the change of the light source. Unfortunately, the reports of these intercomparisons are not available in the ECAC (European Center for Aerosol Calibration) webpage.

The sentence was modified as follow:

“Due to the non-homogeneity of the angular distribution of light intensity of model M9003 (cf. Müller et al., 2009), the light source was changed at SIR in 2013 with the AURORA3000 light source and at CMN in 2009 with an opal glass light source. After the change of the light sources, both nephelometers were examined at the World Calibration Center for Aerosol Physical properties in Leipzig and both performed very well (personal communication from CMN and SIR data providers).”

- 8) Page 8, Line 15-16 – “However, for $SSA < 0.8$ a correction scheme based on particle number size distribution should be used” Was this type of correction necessary for any of the stations? It looks from the table like it may have been performed for some stations, but was it necessary for those stations?**

To the best of our knowledge all stations involved in this investigation applied the Angstrom-based correction scheme to correct the nephelometer scattering data. In the revised version of the manuscript, the sentence was modified as follow:

“These schemes consist of a simple linear correction based on the scattering Ångström exponent (SAE) determined from the raw nephelometer data to take account of the size-distribution-dependent truncation error. It has been demonstrated that these simple correction schemes are accurate for a wide range of atmospheric aerosols and that the uncertainties in the corrections are not expected to be larger than 2% for an aerosol particle population with a single scattering albedos (SSA) greater than 0.8 (Bond et al., 2009).”

- 9) Page 8, Line 19-20 - Only at SIR, FKL, and CMN, σ_{sp} data are not corrected for truncation because σ_{sp} at these observatories was measured at one wavelength. Anderson and Ogren suggest corrections for the nephelometer when the SAE is not available. Doesn't the Mueller scheme also provide an option for correction when no SAE is present? I would expect FKL (a coastal site presumably with large sea salt aerosol) will be quite sensitive to not being corrected for truncation and uncorrected scattering will significantly underestimate the actual scattering.**

During the discussion phase further feedback was needed with data providers regarding the correction of scattering data for some stations. For BIR, MPZ, PUY and TRL the information provided in the metafiles was that scattering data were NOT corrected for truncation, whereas data submitted to EBAS were actually corrected by data providers. Table S1 has been accordingly modified and Figures and Tables changed in order to take into account for this error in the metafiles.

Concerning the Reviewer comment, our opinion is that the correction of the single-wavelength scattering measurements at SIR, FKL and CMN could introduce additional, undesired errors. This is very probably the reason why scattering data provided to EBAS by these three stations were not corrected for truncation.

At CMN and SIR the Ecotech model M9003 was used. Correction schemes for this nephelometer model were provided by Müller et al. (2009). At FKL two nephelometers were used, namely the RR M903 (until 2011) and the Ecotech 1000 (from 2012).

Concerning the Ecotech model M9003, Figure 4 from Müller et al. (2009) shows that the correction factor for this nephelometer model is around 0.97 – 1.00 for SAE around 1.5 – 2.0. The mean SAE at CMN (from TSI measurements performed during 2014-2015; cf. Table S5 in supporting information of this manuscript) was around 2. We do not have an estimation of SAE for SIR observatory which is located in a suburban area 20 km south of Paris. However, the mean SAE at other stations located in West of Europe (PUY, OPE and CBW) was around 1.6-2.0 (after excluding the coastal/marine MHD observatory also located in the Western European sector). Thus, we suppose that the SAE at SIR is probably not low. Thus, the correction factor for both SIR and CMN is close to one. For these reasons we prefer not to correct scattering data from CMN and SIR.

The correction scheme for the RR M903 (deployed at FKL) was also provided by Müller et al. (2009). However, to the best of our knowledge no correction scheme was provided for the Ecotech 1000 which was used at FKL from 2012. Moreover, at FKL the inlet was changed many times (cf. Table 1: whole air (2004-2008), PM10 (2009-2011), PM1 (2011-2012), PM10 (2013-2015)) and the correction factors

provided in literature are also function of the size cut-off used. For these reasons, we prefer not to correct scattering data collected at FKL.

With the permission of the Reviewer, we prefer not to correct the $1-\lambda$ scattering data from CMN, SIR and FKL. Thus, in the revised version of the manuscript all scattering data presented are corrected for truncation with the exception of CMN, SIR and FKL. The following sentence was added at the end of the Paragraph 2.2.2.1:

“Data from the integrating nephelometers used here are corrected for non-ideal illumination of the light source (deviation from a Lambertian distribution of light) and for truncation of the sensing volumes in the near-forward (around 0-10°) and near-backward (around 170-180°) directions (Müller et al., 2009 and Anderson and Ogren, 1998). Correction schemes have been provided by Müller et al. (2009; 2011) for the RR M903 and Ecotech models M9003 and AURORA3000, and by Anderson and Ogren (1998) for the TSI3563. These schemes consist of a simple linear correction based on the scattering Ångström exponent (SAE) determined from the raw nephelometer data to take account of the size-distribution-dependent truncation error. It has been demonstrated that these simple correction schemes are accurate for a wide range of atmospheric aerosols and that the uncertainties in the corrections are not expected to be larger than 2% for an aerosol particle population with a single scattering albedos (SSA) greater than 0.8 (Bond et al., 2009).

The majority of the σ_{sp} data in the EBAS database are corrected for non-ideal illumination and for truncation by the data providers. Exceptions are the scattering data submitted for KOS, MHD, PLA, CMN, FKL and SIR. Scattering data from KOS, MHD and PLA were corrected in this work using the correction scheme provided by Anderson and Ogren (1998) (cf. Table S1 of the Supporting Material). The σ_{sp} data collected at CMN, FKL and SIR are not corrected because the nephelometers deployed at these three stations provide scattering only at one wavelength, thus preventing the estimation of the SAE. Given that the nephelometer correction factors vary as a function of SAE, the assumption of a constant correction factor to correct the $1-\lambda$ scattering data could introduce undesired noise. Moreover, at SIR and CMN, the σ_{sp} is measured with the single wavelength Ecotech nephelometer model M9003 (until 2013 at CMN). The correction curve from Müller et al. (2009; Figure 4) provides a correction factor of around 0.97 to 1.0 for the M9003 for a SAE of around 1.5 to 2. Using the TSI3563 scattering measurements performed at CMN during 2014-2015, we estimated a mean SAE of around 2 for CMN (cf. Table S5). Thus, given the rather small effect of the correction factor estimated for the Ecotech M9003, scattering data from CMN and SIR were not corrected in this work. At FKL the nephelometer models RR M903 (until 2011) and Ecotech 1000 (from 2012) were used (cf. Table 1). To the best of our knowledge, no correction scheme has been provided for the Ecotech 1000. Moreover, at FKL, the inlet was changed many times (cf. Table 1) and the correction factors provided in the literature are a strong function of the size cut-off used. For these reasons, scattering data collected at FKL are not corrected in this investigation.”

10) Page 9, Line 10-11 –“However, the scattering enhancement due to a change in RH between 40% and 50% should be small.” This very much depends on the aerosol type. See hygroscopicity work at several of these ACTRIS sites by Paul Zieger as well as more recent (2017) work on sea salt. How much do the results change if RH<40% is chosen instead of RH<50%? That would (a) be a useful finding and (b) indicate whether the statement above is reasonable (though it would not prove it one way or the other).

In order to take into account the Reviewer comment the following sentence:

“Estimating the aerosol particle light scattering enhancement due to an increase of RH from 40% to 50% is difficult using the data available here because σ_{sp} measurements at RH>40% are not evenly distributed over the measurement periods. In fact, at the majority of the stations RH higher than 40% is registered mostly in summer. However, the scattering enhancement due to the change in RH between 40% and 50% should be small and will not exceed around 3-5% even for more hygroscopic particles (e.g. Fierz-Schmidhauser et al., 2010a,b).”

was removed and replaced with the following sentence:

“Estimating the aerosol particle light scattering enhancement due to an increase of RH from 40% to 50% is difficult using the data available here because the σ_{sp} measurements at a RH>40% are not evenly distributed over the measurement periods, with the majority of the stations registering a RH higher than 40% during the summer. Moreover, the chemical composition of atmospheric aerosol particles is an important factor determining the magnitude of the scattering enhancement due to water uptake, which can then change from one site to another (e.g. Fierz-Schmidhauser et al., 2010a,b; Zieger et al., 2014, 2017). However, the scattering

enhancement due to a change in RH between 40% and 50% should be small and will not exceed few percent even for more hygroscopic particles (e.g. Fierz-Schmidhauser et al., 2010a,b). Table S2 in the Supporting Material reports the percentage of hourly σ_{sp} values collected in the range 40%<RH<50% whereas the frequency distributions of the measured RH are shown in Figure S1.”

Following the Reviewer comment we performed a sensitivity study (Figures reported below) to understand the effect of using data at RH<50 (rather than RH<40%) on aerosol optical properties. However, we confirm what already stated in the manuscript, i.e. that “Estimating the aerosol particle light scattering enhancement due to an increase of RH from 40% to 50% is difficult using the data available here because σ_{sp} measurements at RH>40% are not evenly distributed over the measurement periods. In fact, at the majority of the stations RH higher than 40% is registered mostly in summer.”

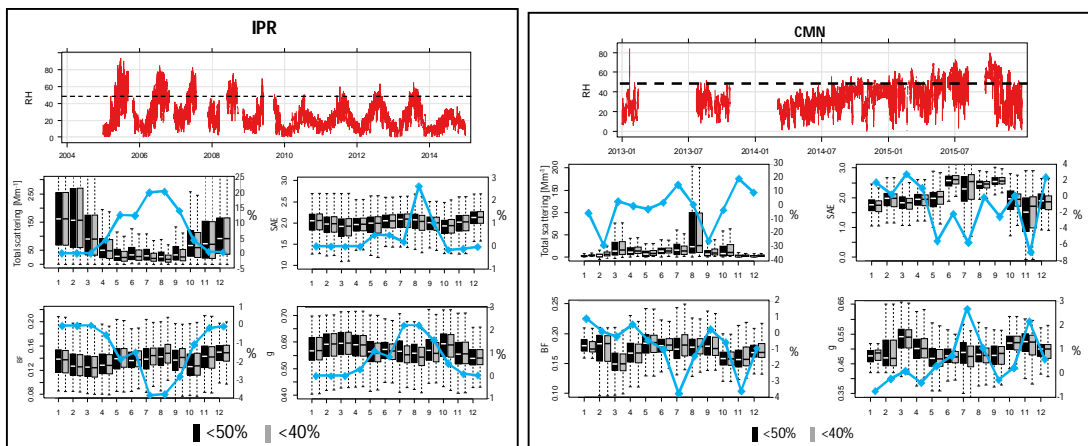
The following figures show the difference between σ_{sp} , SAE, BF and g using data collected at RH<50% and RH<40%. We performed this test for the observatories where the percentage of measurements at RH>50 is high (cf. Table S2), namely: IPR, CMN and SIR. However, we have also performed the same test using two stations (MSA and MPZ) where the RH never exceeded the 40% threshold. For MSA observatory, we compared the measurements performed at RH<40% with those performed at RH<30%. For the MPZ observatory, we compared the measurements performed at RH<30% with those performed at RH<20%. The objective of using MSA and MPZ is demonstrating that the differences in σ_{sp} , SAE, BF and g cannot be only ascribed to differences in RH, but that removing data during specific periods of the year also strongly contributes to the observed differences.

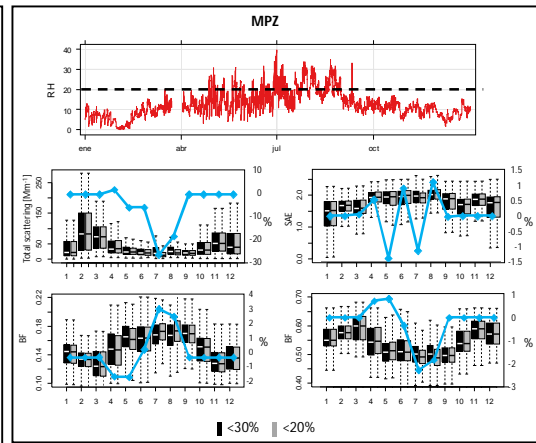
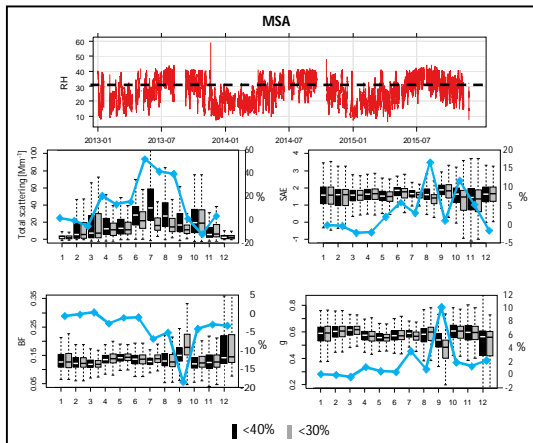
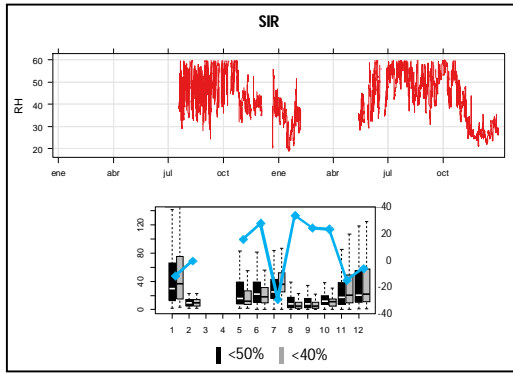
In general, differences up to -30% ÷ +35% were observed for IPR, SIR and CMN for scattering. However, the differences for the intensive optical properties (SAE, BF and g) were much smaller and never exceeding -7% ÷ +3%

The differences (%) are calculated in such a way these are positive when scattering at RH<50% exceeds the scattering at RH<40%. However, for CMN, the difference is negative for example during the month of February and September, thus this difference cannot be ascribed to the scattering enhancement due to water uptake. Rather, this difference is due to the fact that we are removing non-simultaneous data in order to perform the calculations.

Moreover, we observed that also for MSA and MPZ the differences for scattering can be high (-20% ÷ +50%) despite the lower RH taken into account at these two stations.

With the permission of the Reviewer, we would like to use data collected at RH<50% in the revised version of the manuscript. Moreover, we do not think that adding the Figures below to the Supporting Material can add useful information to the reader.





11) Page 9, Line 31 – is 1.5 a reasonable Angstrom exponent for a marine site? Seems a bit high to me.

Surely, a value of 1.5 is probably high for a marine site. In the revised version of the manuscript we provide a range of SAE value and we calculated the scattering at 550 using SAE of 1 and 1.5. The sentence:

” At FKL and SIR, where SAE is not available and assuming a SAE of 1.5, the difference by adjusting to 550 nm is 4.9% at FKL and 26% at SIR, respectively. The higher difference at SIR is due to the fact that measurements at this station are performed at 450 nm.”

Was modified as follow:

“At FKL and SIR, where the SAE is not available, and assuming a reasonable SAE range between 1.5 and 1.0, the difference due to the adjustment to 550 nm is 4.9-3.0% at FKL and 26-18% at SIR. The higher difference at SIR is due to the fact that measurements at this station are performed at 450 nm.”

12) Page 10, Line 28 – I think it would be worthwhile to explain why the blue and red scattering have less coverage. Are the consistent problems with the wavelengths in particular nephelometers or are there data QC issues? (I realize some sites only have green measurements but it's unclear from your statement if those are the sites that don't have red/blue coverage (it wouldn't be expected!) or if it is other sites that do have those measurements but for whatever reason they aren't available. My experience with Ecotech neph data suggests the red wavelength is pretty unstable at many sites within the ACTRIS network and that has implications for SAE climatologies and trends.

As reported in Table S3, the data coverage for scattering at the three wavelengths is very similar at all observatories. Only at CMN the blue and red wavelengths have much lower data coverage because the three wavelength nephelometer was implemented starting from 2014 (only green measured until 2014). At all other observatories the data coverage of scattering is very similar for the three wavelengths.

In order to clarify this point the sentence:

“Exception are σ_{sp} measurements in the blue (450 nm) and in the red (700 nm) and σ_{bsp} measurements at CMN where the three wavelength nephelometer was implemented starting from 2014.”,

was replaced with the following sentence:

“Exceptions are the σ_{sp} measurements at CMN in the blue (450 nm) and red (700 nm) wavelengths which have much less data coverage compared to the green wavelength because the three wavelength nephelometer was implemented at CMN in 2014. Consequently, also the SAE and g have low data coverage at CMN.”

Moreover, we agree with the Reviewer that the red wavelength is especially critical for the Ecotech nephelometer. For this reason, for example, the red was not used at CHC station because of wrong calibrations which effect was especially evident in the red. At all other stations, the red wavelength of Ecotech seemed acceptable. We decided to include the red wavelengths of the majority of Ecotech nephelometers after inspecting the differences between SAE calculated between blue and green and between green and red.

13) Page 12, Lines 4-10 – it would be good to include the physical interpretation of the skewness as well as the mathematical

Following the reviewer comment the sentence:

“Positive skewness is usually observed for positive defined parameters having a frequency distribution with a pronounced right tail indicating the presence of high positive values.”

was replaced with the following sentence:

“The skewness is defined as the third standardized moment of a probability distribution and it is a measure of the asymmetry of the probability distribution. Its value can be positive or negative. Positive skewness is usually observed for parameters which are defined to be positive and it indicates that the tail on the right side of the distribution is longer or fatter than that on the left side. Thus, for a right-skewed distribution, the mass of the distribution is concentrated on the left, and there is a higher probability of measuring a high value compared to a left-skewed distribution. Figure S2 in the Supporting Material shows the frequency and cumulative frequency distributions for σ_{sp} for each station, evidencing the presence of these right-skewed tails.”

14) Page 11, Line 34 - PAL is often considered an Arctic site (see e.g., Backman et al., AMT, 2017) so it may make more sense to group it with ZEP)

Following the Reviewer suggestion the category “Arctic” has been included in the revised version of the manuscript. The PAL and ZEP stations have been included in the new category “Arctic”.

15) Page 14, Lines 19-21 – you have the data and can determine whether the inlet size cut had an effect on the Angstrom exponent. Could plot monthly medians of SAE before and after the inlet was changed.

As suggested by the reviewer we plotted the SAE monthly medians (attached below) before and after inlet change for KPS [PM₁ (2006-04/2008) and PM₁₀ (05/2008-2014)], PAL [PM₅ (2000-08/2005), PM_{2.5} (08/2005-2007) and PM₁₀ (2008-2015)], TRL [whole air (2007-2009) and PM₁₀ (2010-2015)], and MSA [PM_{2.5} (2013-03/2014) and PM₁₀ (04/2014-2015)]. For these stations we calculated the %diff of SAE for different periods: 1) before and after the inlet change (red lines in Figure B), and 2) for periods without inlet changes (black lines in Figure B).

As reported in Figures A and B, the natural variability of aerosol particles over time (black lines in Fig.B) can cause changes in SAE of the same order of those possibly caused by the inlet change (red lines in Fig.B).

Thus, it is very difficult to evaluate the effect of the inlet change on SAE given that measurements performed with different cutoffs are not simultaneous. For this reason, we also report in the figures below the same analysis for MPZ and MSA where the inlet was not changed during the sampling period. The aim of reporting the comparison of SAE for different periods for MPZ and MSA is confirming that natural variability also can considerably affect the variability of SAE.

At MSA the %diff of SAE between the two selected periods is similar to the %diff observed for PAL or TRL (before and after the inlet change). At MPZ the %diff is smaller suggesting more homogeneity in some microphysical properties of atmospheric particles such as size (cf. Figures S4 in Supporting Material). From Fig. S4 we can say that also at KPS more homogeneity in particles size can be expected (very narrow SAE frequency distribution as for MPZ). In fact, at KPS the %diff is rather small compared to the other stations.

In conclusion, we think that the analysis of SAE before and after the inlet change cannot be used to determine how much the inlet change affected the calculated SAE.

Moreover, the analysis of the trends of scattering and SAE for PAL has been already reported in literature for the period 2000-2010 (Collaud Coen et al., 2013). Furthermore, Lihavainen et al. (2015a) assumed that the inlet changes at PAL had only minor effects on scattering, because the number concentration of coarse particles is very low at this observatory.

In conclusion, we do not think that adding the Figures below to the Supporting Material can help understanding the effects of the inlet change. Moreover, given the huge amount of data provided by many of the stations involved in this investigation, some latitude in data processing amongst stations should be deemed acceptable. Thus, despite the possible effect of the inlet changes, a detailed picture of scattering properties of surface aerosol particles is clearly provided with the data used here.

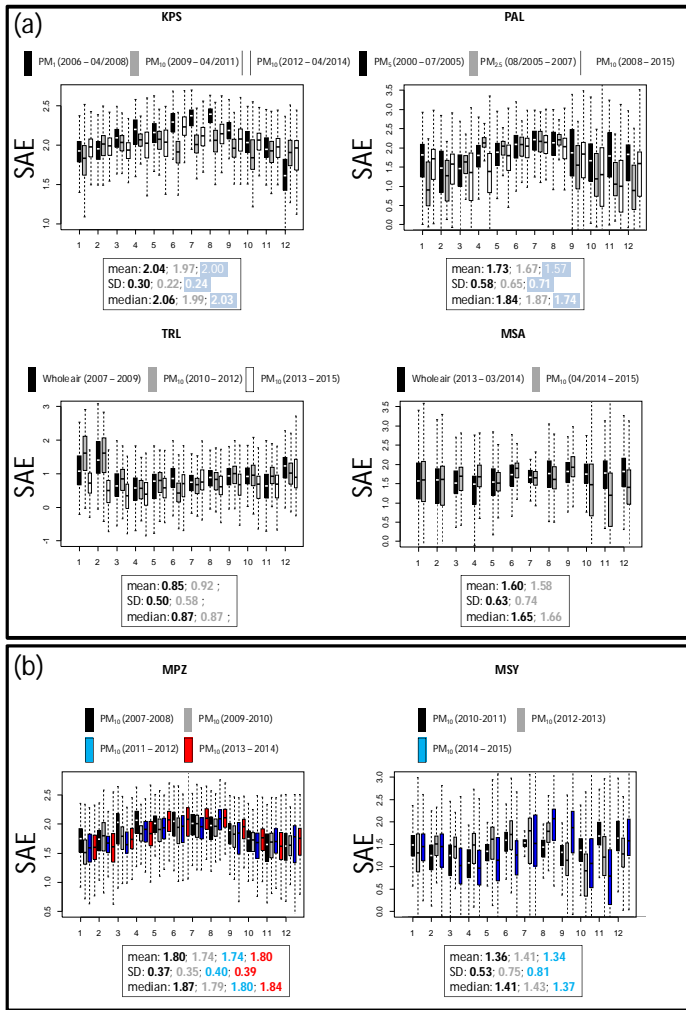


Figure A

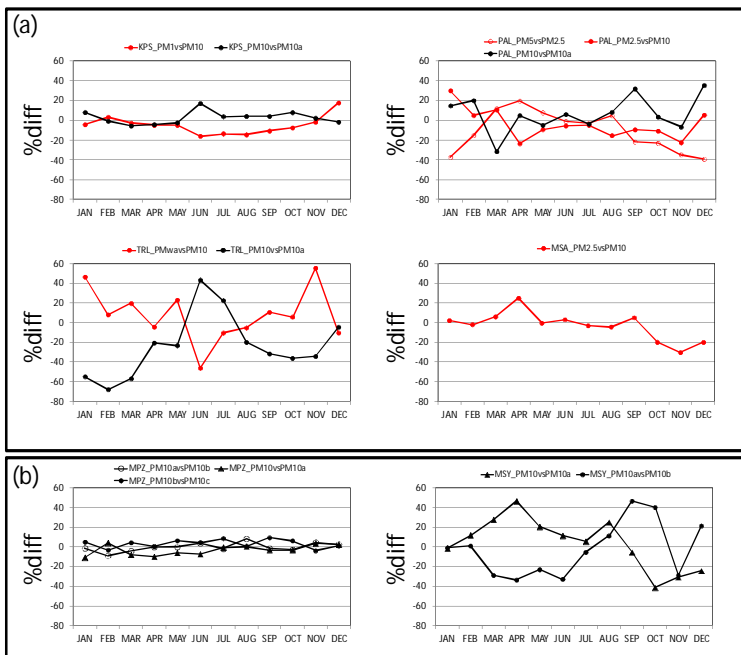


Figure B

16) Page 15, Line 22 – why do BIR and PLA have a large particle peak?

These peaks with low SAE at BIR and PLA are also probably due to the presence of marine aerosols at these sites.

The sentence was modified as follow:

“BIR and PLA also show an enhanced left peak in the SAE frequency distributions likely due to the presence of coarse marine aerosols at these sites.”

17) Page 20, Lines 20-26 – these percentage comparisons are misleading – scattering Angstrom exponent is constrained between the values of approximately 0 and 3 and therefore you will not see anywhere close to the same percentage change as you would see for scattering which is unconstrained. I would remove lines 20-26.

Yes. We agree. The sentence was removed in the revised version of the manuscript.

18) Page 25, Line 32 – PAL trends in SAE – you may want to discuss with John Backman – the Level 2 PAL data in EBAS has some suspicious red values for several years.

Yes. It is true that Level 2 EBAS PAL data has some suspicious red values. We detected such problem during the double check we performed on the data. In the manuscript we already removed data collected at PAL during: the whole year 2017; July and August 2002; September and October 2003; May-June-July 2015.

19) Page 27, Line 29 – are there day/night differences at coastal sites due to onshore/offshore flow?

Onshore/offshore flows surely have important effects at coastal sites. However, we do not know if these flows have any effect at the mountain sites considered for the day/night analysis presented in the manuscript.

Editorial comments:

20) It would be good to have a native English speak read and edit the paper before resubmission.

Following the Reviewer suggestion, the manuscript has been revised and edited by a native English speak.

21) It would also be really helpful to the reader to organize the discussion better – the paragraphs are really long and it's occasionally hard to follow the arguments because of that. I've suggested some places where the really long paragraphs could be broken into smaller paragraphs but some transitional sentences may be needed.

Following the Reviewer suggestions, smaller paragraphs were added in the revised version of the manuscript

22) Page 4

Break into smaller paragraphs: line 11 – start new paragraph, line 19 – start new paragraph, line 32 – start new paragraph

Done

23) line 15 – there are evidences -> there is evidence

Done

24) line 17 – phrasing of ‘would eventually unmask the global warming’

Following the Reviewer comment, the sentence was modified as follow:

“In fact, there is evidence suggesting that the observed (and projected) decrease in emissions of anthropogenic aerosol particles in response to air quality policies will eventually exert a positive aerosol effective radiative forcing at the top of the atmosphere (Rotstayn et al., 2013). Thus, current emission controls could both enhance climate warming while improving air quality (e.g. Stohl et al., 2015).”

25) line 22-23 – phrasing of ‘Several international projects are providing in the last decades important information on the atmospheric particle properties worldwide’

Following the Reviewer comment, the sentence was modified as follow:

“In recent decades, several international projects have provided important information on atmospheric particle properties worldwide.”

26) line 25 completed -> complemented

Done

27) line 26 - 27 USA or EMEP -> USA and EMEP

Done

28) line 30 – define RTD

The sentence has been modified as follow:

“... and from short-term RTD (Research and Technological Development) projects such as EUCAARI (European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions; <http://www.cas.manchester.ac.uk/resprojects/eucaari/>).”

29) Line 36 – You should be careful here. (1) EBAS also includes data from the IMPROVE network nephelometers which are operated outside at ground level and at ambient conditions with no size cut. These IMPROVE data aren’t really comparable to the ACTRIS data sets discussed here. (2) Additionally there are other sites making nephelometer measurements that aren’t providing the data to EBAS (see comment below for page 5, lines 20-22).

We agree with the Reviewer that ACTRIS and IMPROVE nephelometer data are not really comparable. The following sentence has been added:

“However, EBAS also includes data from the IMPROVE network nephelometers, which latter are operated at ambient conditions with no size cut, as a result of which these IMPROVE data are not directly comparable to the ACTRIS dataset discussed in this investigation.”

Page 5

30) Line 7 ‘that decreasing or’ -> that a decreasing or

Done

31) Lines 13-22 – should cite Sherman et al 2015 – it is an updated version of Delene and Ogren 2002

The reference Sherman et al. (2015) was added to the text and the bibliography.

32) Lines 20-22 – this statement only relates to multi-station studies although Andrews et al included 3 mountain sites in Asia (WLG (China), PYR (Nepal), LLN(Taiwan)). Please rephrase to make clear that there are measurements outside of Europe and the US, but that many of those measurements have primarily been written about in isolation (e.g., not in the context of other sites). For example, FMI has reported on long-term optical properties in Saudi Arabia and South Africa. Paolo Laj et al have reported on aerosol optical properties in Nepal, and Paolo Artaxo in Brazil. Note: this manuscript doesn’t really change this since it is focused on European sites as well and I think you should remove the non-European sites you do include to improve the discussion and flow.

We agree with the Referee that this manuscript doesn’t really change the fact that all the multi-sites studies presenting aerosol particle optical properties mostly focus on Europe and US. For this reason the following sentence:

“Thus, the number of papers reporting aerosol particle optical properties measured at different sites is rather scarce and unfortunately almost inexistent outside Europe and the United States.”

was removed in the revised version of the manuscript.

Moreover, as suggested by the Reviewer, the non European sites have been removed from the manuscript and presented mostly in the Supporting Material.

33) Line 26 - ‘related with aerosol phenomenology’ -> related to aerosol Phenomenology

Done

34) Line 31 – ‘Zanatta’ -> ‘and Zanatta’

Done

Page 6

35) Lines 1-13 should be in methods section 2.2.3, not introduction

Lines 1-13 have been moved to the Section 2.2.3, as follow:

“2.2.3 Calculation of aerosol particle intensive optical properties

Starting from the spectral σ_{sp} measurements performed at the ACTRIS observatories, three intensive aerosol particle optical parameters can be estimated, namely; the scattering Ångström exponent (SAE), the backscattering fraction (BF) and the asymmetry parameter (g). These intensive properties do not depend on the PM mass concentration and are directly related to aerosol particle properties such as size, shape, size distribution and chemical composition. The SAE can be considered as a proxy for the aerosol particle size range with a higher (lower) SAE associated with predominance of fine (coarse) aerosol particles (e.g. Seinfeld and Pandis, 1998; Esteve et al., 2012; Valenzuela et al., 2015 among others). The BF and g parameters are calculated quantities that influence the variability of the radiative forcing efficiency and that represent the angular light scattering of aerosol particles. For computational efficiency, the angular light scattering is often represented by a single value (BF, σ_{sp}/σ_{bsp} or g) (Andrews et al., 2006).”

36) Line 1 – delete ‘In fact,’

Done

37) Line 1 – add the word ‘spectral’ in front of σ_{sp}

Done

38) Line 5 – particles -> particle

Done

39) Line 7 – associated to -> associated with

Done

40) Line 7 - course -> coarse

Done

41) Line 11 – a better reference is the Andrews et al. 2006 reference which you also cite later. Ogren 2006 is gray (not peer-reviewed) literature and the Andrews paper is the peer-reviewed version of it

Done

42) Line 18 – performed -> characterized

Done

43) Line 19 – observatories -> observatory

Done

44) Line 19 – measurements -> measurement

Done

45) Line 20 – divided in -> divided into

Done

46) Line 22 – coastal – how close to the sea coast?

The sentence has been modified as follow:

“coastal: includes observatories located close to the coast (<1-4 km);”

47) Page 6 -lines 26-36 and Page 7 lines 1-4 - Delete and say the categories for each station are given in table 1. No need to have in paragraph form also.

Done

Page 7

48) Line 5 - Earlier you say mountain sites are higher than 1km, but then you characterize HPB at 985 m as a mountain. Should change previous statement and say mountain sites are at or higher than 985 m to be consistent.

The sentence has been modified as follow:

“mountain: includes those observatories located at more than 985 m above sea level (the lowest altitude among the mountain observatories included here);”.

49) Line 9 – investigation -> investigations

Done

50) Lines 11-15 – delete - you say this in the trends section

Done

51) Line 25 - ‘Most used nephelometer models are the’ -> The most common nephelometers in the ACTRIS program are the

Done

52) Line 27 - ‘Other used models’ -> Other models used

Done

53) Line 37 – ‘guarantee the quality and comparability of the data.’ This is a strong statement. There are RH control issues, wavelength, differences, data QC issues and many of the systems operate with different inlets/size cuts (e.g., whole air, pm2.5 pm 10). Do stations have different procedures for dealing with negative values close to zero? How do they deal with local contamination? There are unfortunately a lot of issues and while the nephs themselves are probably quite comparable when they are operated side-by-side as happens at a Leipzig workshop the operating conditions at the stations will vary and make the measurements less comparable. What happens if a nephelometer fails a performance check – is the data prior to that time nvalidated? You discuss some of these issues below in the sections below but I recommend changing the text as follows:

{Recommended quality assurance procedures during on-site operation as described in GAW (WMO/GAW, 2016), guarantee the quality and comparability of the data. Moreover, most of the integrating nephelometers involved in ACTRIS have undergone performance checks at scheduled times at the World Calibration Center for Aerosol Physical properties of ACTRIS/GAW.}

Change text in red to text in blue •

{Recommended quality assurance procedures during on-site operation as described in GAW (WMO/GAW, 2016), help to ensure the quality and comparability of the data. Additionally, most of the integrating nephelometers involved in ACTRIS have undergone performance checks at scheduled times at the World Calibration Center for Aerosol Physical properties of ACTRIS/GAW. 2.2.2 Data treatment The σ_{sp} and σ_{bsp} data reported to EBAS and used in this work are referenced to standard T (273.15 °C) and P (1013 hPa) conditions. There are however station-to-station differences (e.g., sizecut, RH control, wavelength, data processing, etc) which are addressed in the sections below. 2.2.2.1}

Following the Reviewer comment the sentence was changed as follow:

“Recommended quality assurance procedures during on-site operation, as described in GAW (WMO-GAW Report, 2016), help to ensure the quality and comparability of the data. The nephelometers included in this investigation are regularly calibrated using span gas and are zero adjusted using particle-free air. Additionally, most of the integrating nephelometers employed in ACTRIS have undergone a schedule of performance checks at the World Calibration Center for Aerosol Physics of ACTRIS/GAW.”

“2.2.2 Data treatment

Data used in this investigation include hourly averaged Level 2 aerosol particle scattering data downloaded from the ACTRIS/EBAS Data Centre web portals (www.actris.nilu.no; www.ebas.nilu.no; last downloads August 2017). The σ_{sp} and σ_{bsp} data reported to EBAS and used in this work are referenced to standard T (273.15 °C) and P (1013 hPa) conditions. Data consistency is critical when comparing many years' worth of data from different stations. In this work, the Level 2 scattering data were further reviewed in order to ensure a high quality of the data presented. There are however station-to-station differences (e.g. sizecut, RH control, wavelength, data processing, etc.) which are addressed in the sections below.”

Page 8

54) Line 30 – should say here in the first sentence that the GAW (and ACTRIS?) protocol is RH<40%. You say it later in the paragraph but it should be at the very beginning of the paragraph.

Done

Page 9

55) Lines 14-16 - this should be moved elsewhere – it is not related to the RH discussion. See my suggestion above which puts it after section heading 2.2.2

The sentence was moved to the beginning of the section 2.2.2.

56) Line 20 – other used wavelengths -> other wavelengths used

Done

57) Line 23 – most used σ_{bsp} most common

Done

58) Line 24 – following Sections -> following sections,

Done

59) Line 27 – (and at CMN -> (or at CMN

Done

60) Line 28-29 - measured at different wavelength than 550 nm -> measured at additional wavelengths to 550 nm

Done

Page 10

61) Line 6 - delete (with $\lambda_1 > \lambda_2$): this is not a necessary condition for the SAE equation you provide - you can flip things around so long as you are consistent, e.g., if $ssp_{550}=40$ and $ssp_{700}=30$, then $SAE = -\log(30/40)/\log(700/550) = -\log(40/30)/\log(550/700) = 1.19$

Done

62) Line 8-9 – ‘Here, the SAE is calculated as linear estimation of σ_{sp} measured at the three available wavelengths.’ This statement is inconsistent with the equation describing Angstrom exponent (eq 1). Further the relationship is not linear – that’s why the equation has logs in it. Please clarify what was meant.

The sentence was changed as follow:

“Here, the SAE is derived from a multispectral log linear fit based on the three nephelometer wavelengths.”

63) Line 14 – given radiation -> given direction

Done

64) Line 18 – see previous comment about the Ogren 2006 citation

Done

Page 11

65) Section 3.1 – two points: (a) Why no separate section for Arctic sites? Could move discussion of ZEP and PAL to an arctic section (as suggested above I would remove discussion of TRL from main manuscript). (b) Why not order the discussion of sites from typically cleanest (arctic) ->mountain ->coastal ->...-> to typically dirtiest (urban). That would make the discussion easier to follow I think.

We agree with the Reviewer. A separate section for Arctic sites (ZEP and PAL) has been introduced in the revised version of the manuscript. The TRL station (together with CHC) was moved in the Supporting material. Sections 3.1 and 3.5 were modified accordingly to the Reviewer comment with the discussion presented from cleanest sites to urban sites. Figures and Tables were also accordingly modified.

66) Line 20 - placements -> locations

Done

67) Line 32 – because their -> because of their

Done

68) Line 35 – since these are -> because they are

Done

69) Line 27 – low scattering are -> low scattering is (or low scattering value are)

Done

70) Page 11, line 16 – page 12, line 10 – Split the paragraph into several smaller paragraphs: Start a new paragraph at page 11 line 25 for discussion of figure 3. Start a new paragraph at line 29. Start a new paragraph page 12 line 4.

Done

71) Lines 29 to page 12, line 4 – This discussion is hard to follow – in part because the site types as defined earlier aren't used consistently.

Following the Reviewer comments the presentation of the results is organized differently in the revised version of the manuscript. The sentence (from pag. 11, line 29 to pag.12, line 4) was moved at the beginning of section 3.1 as follow:

3.1 Variability of σ_{sp}

Figure 2 shows the box-and-whiskers plots of σ_{sp} measured at the stations included in this investigation. In Figure 2, the observatories are grouped based on their placement and ordered according to their geographical location. Table S4 and Figure S2 in the Supplementary Material report, respectively, the statistics of σ_{sp} (mean, standard deviation, minimum and maximum values and 5th, 25th, 50th, 75th, and 95th percentiles) and frequency and cumulative frequency distributions.

In each geographical sector, an increasing gradient of σ_{sp} is generally observed when moving from mountain to regional and to urban sites. Thus, the σ_{sp} values measured at mountain sites are lower than the measurements made at other locations (coastal to urban) even if exceptions are observed in some sectors.

A large range of σ_{sp} coefficients is observed across the network, ranging from median values lower than 10 Mm⁻¹ to values higher than 40 Mm⁻¹. Overall, the lowest σ_{sp} is on average measured at remote stations because of either: a) their altitude, for example JFJ is located in Central Europe at more than 3500 m a.s.l. and CHC in Bolivia is at around 5300 m a.s.l. (cf. Figure S3), or b) because of their large distance from pollution sources, for example the Arctic ZEP and PAL stations, TRL station (cf. Figure S3) and some regional sites in the Nordic and Baltic sector such as BIR and SMR. Higher σ_{sp} values (medians > 40 Mm⁻¹) are on average registered at more polluted sites, such as some urban sites in Southern Europe (UGR and DEM), some regional sites in Eastern and Central Europe (KPS and IPR, respectively) and one coastal site in the Nordic and Baltic sector (PLA). The observed variation is consistent with the differences in particulate matter (PM) mass concentrations, PM chemical composition, particle number concentration and absorption coefficients observed across Europe, as described for example by Putaud et al. (2010), Asmi et al. (2011) and Zanatta et al. (2016)."

Page 12

72) Line 13 – placements ->environments

Done

73) Line 30 – here you combine regional and continental stations for discussion – I do think it makes sense to call them clean and polluted continental stations – it would make the discussion simpler.

Following the Reviewer comments the PAL station is included among the Arctic stations together with ZEP. Thus, the only other continental station included in this work is VHL. For this reason, the category continental was removed in the revised version of the manuscript. The category Regional/rural was added/modified.

See also the reply to the Reviewer comment #4: “We thank the Reviewer for this comment which considerably simplifies the discussion. As suggested by the Reviewer the PAL station was included in the “Arctic” category in the revised version of the manuscript. Thus, following the EBAS definition, the only continental station is VHL. Consequently, the category “continental” was removed in the revised version of the manuscript and VHL was included in the new “regional/rural” category.”

74) Line 31 – present -> exhibit

Done

75) Line 33-34 – I think ‘linked to strong stable air with thermal inversion’ this could be better phrased: linked to stable air due to strong thermal inversions

Done

76) Line 33-34 – delete ‘On the other side,’

Done

Page 13

77) Line 2 – instead of continental should say Nordic/Baltic continental sites because those are the subset of sites VHL is being compared to rather than all continental sites.

In the revised version of the manuscript the category “continental” was removed and the continental VHL station included in the new category “Regional/rural”. Please, see reply to Reviewer comment #4.

Page 14

78) Section 3.2 – maybe would be easier to read if had subsections by station type or could have two subsections: (a) by geography (east to west) (b) by type (coastal, mountain, etc). Right now they are a bit intertwined.

Following the Reviewer suggestion two subsections were added to the section 3.2, namely:

- 3.2.1 Variability of SAE by geographical sector, and
- 3.2.2 Variability of SAE by station type

79) Line 16 – start new paragraph

Done

80) Line 37 – the SAE data -> the frequency plot of the SAE data

Done

Page 15

81) Line 13 – start new paragraph

Done

82) Line 18 – cite work by Zieger et al 2010 at ZEP (their figure 4) which shows presence of sea salt at ZEP.

Done

83) Line 24 – ‘Differently than σ_{sp} ,...’ -> Unlike σ_{sp} , ...

Done

84) Line 34 – start a new paragraph with ‘Also at...’

Done

Page 16

85) Section 3.3 – maybe would be easier to read if had subsections by station type or could have two subsections: (a) by geography (east to west) (b) by type (coastal, mountain, etc). Right now they are a bit intertwined.

Following the Reviewer suggestion two subsections were added to the section 3.2, namely:

3.3.1 Variability of g by geographical sector, and

3.3.2 Variability of g by station type

86) Line 14 - start new paragraph

Done

87) Line 14 – “..thus the lower BF the higher is g ..” • change to “...with lower BF corresponding to higher g ..”

Done

88) Line 16-17 – “Higher g median values are in some cases observed at mountain sites compared to regional or urban environments” -> At some mountain sites higher median g values are observed relative to the g values obtained at regional or urban locations.

Done

89) Line 18 – “...European sector or HPB...” -> ...European sector and for HPB...

Done

90) Line 19 – “However, exceptions are observed for example for CMN...” -> However, exceptions are observed. For example, at CMN, ...

Done

91) Line 23 – start a new paragraph

Done

92) Line 35 – start a new paragraph

Done

Page 17

93) Line 10 – start a new paragraph

Done

94) Line 10 – “Moreover, the refractive...” • The refractive ...

Done

95) Line 13 – “...did non linearly...” • ...non-linearly decreased...

Done

96) Line 16 – higher -> larger

Done

97) Line 17 – “On the other side, Obiso et al. (2017) showed...” • Obiso et al. (2017) also showed...

Done

98) Line 20 – “This kind of...” • These kind of ...

Done

99) Line 36 – retitle section BF and g vs scattering relationships. Both are talked about in the section so the title is a little confusing

Done

Page 18

100) Line 7 – Sherman et al., ACP 2015 also www.atmos-chem-phys.net/15/12487/2015/

The reference has been added to the main text and to the bibliography.

101) Line 37 “or higher 1.5” • or higher than 1.5

Done

102) Page 18, Line 20 – page 19, line 17 – this paragraph should be better organized and split into two smaller paragraphs. It’s a bit hard to follow in its current form.

Please, see reply to the comment below (#103)

103) Section 3.4.1 – general comment – I would recommend first discussing the SAE/scattering relationships and then relating it to the g/BF relationships. Move lines 20-23 to a paragraph at the end of the section.

Also, perhaps it would make sense to split this section into aerosol types ‘marine/dust’ and ‘anthropogenic’.

Reply to comments #102 and #103: With the permission of the Reviewer we would like to discuss first the g -scattering relationship and then the SAE-scattering relationship. This because the g -scattering relationships are very similar at all sites. The g always increases with increasing σ_{sp} . Then, we comment the variations of SAE-scattering relationships among the observatories in function of the g -scattering relationships.

Moreover, we do not find a clear way to split the “SAE- σ_{sp} relationships” section. Surely, the presence of coarse dust aerosols or sea salt can lead to a reduction of SAE with increasing σ_{sp} . This is observed for example at IZO and PLA observatories which are affected by dust and sea salt, respectively. However, the same SAE vs. σ_{sp} relationship is not observed at MHD observatory which is also affected by sea salt. Moreover, the reduction of SAE with increasing σ_{sp} is also observed at sites not affected by dust such as CBW, KPS or MPZ for example.

We think that the analysis of the aerosol particles size distribution measured at the different observatories could probably help in properly separating the section. However, presenting the aerosol particle size distributions is far behind the scope of the present work.

In order to improve the readability of the section, the section was divided in different paragraphs.

Page 19

104) Section 3.5 – my personal suggestion would be to switch the order of sections 3.4 and 3.5. Sections 3.1-3.3 talk about overall variability of the different parameters on an annual basis. It seems logical to talk next about the seasonal variability and tie it into annual variability. Once the individual parameter variability has been discussed then it makes sense to look at how those parameters co-vary (i.e., 3.5). Note: that's also the order those topics are presented in the conclusions.

As suggested by the Reviewer the order of the sections 3.4 and 3.5 was changed in the revised version of the manuscript.

105) Line 25 - ad -> and

Done

Page 20

106) Line 6 – start new paragraph

Done

107) Lines 11-13 – “At the southern station of MSA the observed less pronounced seasonal cycle of SAE could be related with the Saharan dust outbreaks which contrast the PBL transport of fine particles observed at other mountain sites” it's unclear what is being said here; is MSA not impacted by dust?

As suggested by the Reviewer the following sentence:

“At the southern station of MSA the observed less pronounced seasonal cycle of SAE could be related with the Saharan dust outbreaks which contrast the PBL transport of fine particles observed at other mountain sites.”

was replaced with the following sentence:

“At MSA in Southwestern Europe, the observed less pronounced seasonal cycle of SAE could be due to the contribution of Saharan dust in spring/summer, which contrasts with the PBL transport of fine particles observed at other mountain sites during the warm season.”

108) Line 14 - "July-August being the Saharan dust outbreaks very" · July-August. The Saharan dust are very...

The sentence was modified as follow:

"At IZO, the SAE reaches its lowest values during July-August in conjunction with the peak frequency of dust events (Rodríguez et al., 2015)."

Page 21

109) Line 7 - "being the SAE" · with the SAE

Done

Page 22

110) Split the southern Europe paragraph into several paragraphs. Line 15 -start new paragraph. Line 27 start new paragraph.

Done

Page 23

111) Line 3 - citation for recommendation about having more than 10 years of data for trend analysis?

This recommendation was based on a recent work from the Task Force on Measuring and Modelling (TFMM - CLRTAP). In this work the Chemical Coordinating Centre (CCC; <http://www.nilu.no/projects/ccc/>) presented a sensitivity study on Mann Kendall (MK) test demonstrating that the chances that the MK methodology detects a significant trend in a time series with 11 years of data are very small when the actual trend is of the order of 1 %/y. Furthermore, they show that with a trend of 2 %/y and even 3 %/y one could only be fairly certain to find a trend when the natural (inter-annual) variability is small (5 %) [https://wiki.met.no/_media/emep/emep-experts/mannkendall_note.pdf.]

However, this sensitivity study was performed analyzing 11yr and 23yr of data which were the periods selected for the EMEP Report 1/2016 on air pollution trends in the EMEP region between 1990 and 2012 (Colette et al., 2016; http://www.unece.org/fileadmin/DAM/env/documents/2016/AIR/Publications/Air_pollution_trends_in_the_EMEP_region.pdf).

Thus, this sensitivity study was not meant to provide general recommendations.

For this reason we prefer to remove the sentence from the manuscript.

112) Line 20 - start new paragraph

Done

Page 24-25

113) This section (3.6.1) needs to be broken into smaller paragraphs! Line 9 - start new paragraph. Line 17 start new paragraph.

Done

114) Line 14-15 - "A statistically significant decreasing trend of asp at IPR was also reported by Putaud et al. (2014) for the period 2002 - 2010."-> delete from text and put as footnote to Table 2.

Done

- 115) **Line 16-16 – “As reported in Table 2 statistically significant decreasing trend for asp is observed at around 50% of the stations considered here.” -> delete from text – put the 50% number in the first sentence of the first paragraph on line 2 of this page.**

Done

Page 25

- 116) **Line 8 – start new paragraph**

Done

- 117) **Line 36 – start new paragraph**

Done

Page 26

- 118) **Line 9 start new paragraph about BF trends**

Done

- 119) **Line 21 start new paragraph**

Done

Tables and Figures

- 120) **Table 1 – Explain that the ‘observatory code’ is ACTRIS’ code (or EBAS?) (or GAW?). Note where necessary any differences between GAW ids and station ids (e.g., Finokalia’s GAW id is FIK and there may be others).**

The following sentence was added as footnote in Table 1:

(1) Observatory codes from EBAS; (2) GAW code: FIK; (3) GAW code: VAV; (4) GAW code: CES; (5) GAW code: MEL;”

- 121) **Figure 2 – you could put this on log scale**

Done

- 122) **Figures 2, 4 and 5 – how would this look if you had panes for different site types and then organized by geographical region? For example, you would have a pane for mountain sites and then sections for Nordic (empty), western (puy), central (jfj, cmn, hpb), etc. I think doing that would make it easier to see the east west shift and also commonalities among site types. You could keep the boxes colored by geographic region within each pane. You could do a similar thing with figure 6.**

Following the suggestion of the Reviewer Figures 2, 4, 5 have been modified. In the revised version of the manuscript the panes report different site types (Arctic, mountain, coastal, regional/rural and urban/sub-urban) and the boxes are colored in order to distinguish among the different geographic regions. For simplicity the ZEP station was included among the Nordic and Baltic stations.

Figure 6 and Figures 7, 8, 9 and Table 1 have been modified accordingly to the new selection of station categories.

Moreover, Figures and Tables in the Supporting material have been also accordingly modified. All Tables were modified the observatories ordered as a function of the station setting: from Arctic to Urban/sub-urban sites.

Supplemental materials

- 123) Table S1 – how is this table organized? It's not alphabetical or by geography. Or by instrument type.**

The table was organized in alphabetical order in the revised version of the manuscript.

- 124) Table S2 - should provide number of points with RH>40% for each station (i.e., how many points are above the GAW and ACTRIS protocol value) as well as the number of points with RH>50%. Caption should state that table is organized by decreasing number of points with RH>50%.**

Following the Reviewer comment we changed the Table S2 in the revised version of the manuscript so that it presents the percentage of σ_{sp} data (not RH data) collected at $40\% < RH < 50\%$.

In the ACPD version of the manuscript, the Table S2 reports the percentage of RH data higher than 50%. This information is misleading given that often σ_{sp} data collected at $RH > 50\%$ (or 40%) are removed by data providers or flagged as non valid because of instrument failure, calibration periods, unspecified contamination or local influence, etc.

Nevertheless, the % of σ_{sp} data collected at $40\% < RH < 50\%$ is not small at some stations. This is clearly represented in Table S2.

In the table S3 in supporting material we report the percentage [%] of data coverage at the 28 ACTRIS stations included in this study. Percentages are calculated as the ratio between the number of scattering (backscattering) data used in this investigation and the total number of hours during the sampling period at each station. Removed data include data flagged as non valid by data providers (instrument failure, calibration periods, unspecified contamination or local influence, etc) or obtained at RH higher than 50%.

Consequently, in the revised version of the manuscript the Table S2 in supporting material reports the number of σ_{sp} hourly data used in this investigation and the % of hourly σ_{sp} collected at $40\% < RH < 50\%$.

The text in the manuscript describing Table S2 has been accordingly modified.

- 125) Figure S1 – should make the x-axes cover the same range (0-80%?) and draw a vertical line at 40% (GAW protocol value) and 50% (value chosen for this paper). Lines could be added indicating the median RH distribution as a function of season. Explain why different widths of bars on distribution plots (presumably because different numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.**

We changed the Figure S1 in the revised version of the manuscript to include vertical lines for 40% RH, 50% RH, and medians RH values by season. X-axes was set to 0-80%. Consistent widths of bars are now used.

- 126) Table S3 – state in caption what colors λ_1 , λ_2 , and λ_3 correspond with. (not the wavelength because that obviously changes with instrument and time period)**

Done

- 127) Figure S2 – remove map – that's already a figure in the manuscript. Explain why different widths of bars on distribution plots (presumably because different numbers of**

data points). I think it would make more sense to use consistent widths for the distribution plots.

Figure was modified so that the width of the bars is equal for all stations. The map was removed. Colors were also added to represent the geographical location of each station.

128) *Figure S3 – remove map – that’s already a figure in the manuscript. Explain why different widths of bars on distribution plots (presumably because different numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.*

Figure was modified so that the width of the bars is equal for all stations. The map was removed. Colors were also added to represent the geographical location of each station.

129) *Figure S4 – remove map – that’s already a figure in the manuscript. Explain why different widths of bars on distribution plots (presumably because different numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.*

Figure was modified so that the width of the bars is equal for all stations. The map was removed. Colors were also added to represent the geographical location of each station.

130) *Figure S6b – explain the different colors of dots in the figure caption. Perhaps you could make the bars in figure S6a the same color as the dots in figure S6b and then use some other color for the dots in figure S6a*

Colors were added to the figure to represent the geographical location of each station.

131) *Figure S8 – has MTC instead of CMN*

Done. Moreover, the figure was modified and the color codes for different geographical locations were added.