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

GENERAL REMARKS

The manuscripts approaches a European aerosol phenomenology on scattering properties of atmospheric aerosol particles from 28 ACTRIS sites, based on measurements with various types of integrating nephelometers. The manuscript focuses exclusively on ACTRIS sites in Europe, with the addition of Arctic and Antarctic stations and one mountain station in Bolivia, operated jointly by ACTRIS and local partners. The efforts for bringing this extensive data set together are huge and the richness of data is the major contribution of this work to an important scientific discussion on the long-term evolution of extensive and intensive aerosol properties in industrialized and rural regions of the world.

However, in its current version, the manuscript is very difficult to read and it is hard to follow any story line of the paper. Major revisions are required to make this manuscript acceptable for publication in ACP.

Major concerns refer to the organization of the manuscript, presentation of results and lacking of joint in-depth analyses of the observations in combination with existing publications on the long-term evolutions of aerosol optical properties.
Before resubmission of the manuscript, the following concerns should be considered:

Organization of the manuscript and the presentation of results:

1) The abstract is far too long and requires substantial shortening.

Following the Reviewer comment, the Abstract (reported below) was slightly shortened in the revised version of the manuscript. However, given the amount of information provided by this work, it was difficult to further shorten the Abstract. Now the Abstract is less than one page (28 lines, 376 words).

"Abstract

This paper presents the light scattering properties of atmospheric aerosol particles measured over the past decade at 28 ACTRIS observatories which are located mainly in Europe. The data include particle light scattering (σsp) and hemispheric backscattering (σbsp) coefficients, scattering Ångström exponent (SAE), backscatter fraction (BF) and asymmetry parameter (g). An increasing gradient of σsp is observed when moving from remote environments (Arctic/mountain) to regional and to urban environments. At regional level in Europe, σsp also increases when moving from Nordic and Baltic countries and Western Europe to Central/Eastern Europe whereas no clear spatial gradient is observed for other station environments. The SAE does not show a clear gradient as a function of the placement of the station. However, a West to East increasing gradient is observed for both regional and mountain placements suggesting a lower fraction of fine-mode particle in Western/Southwestern Europe compared to Central and Eastern Europe where the fine-mode particles dominate the scattering. The g does not show any clear gradient by station placement or geographical location reflecting the complex relationship of this parameter with the aerosol particles physical properties. Both the station placement and the geographical location are important factors affecting the intra-annual variability. At mountain sites, higher σsp and SAE values are measured in the summer due to the enhanced boundary layer influence and/or new particles formation episodes. Conversely, the lower horizontal and vertical dispersion during winter leads to higher σsp values at all low altitude sites in Central and Eastern Europe compared to summer. These sites also show SAE maxima in the

summer (with corresponding g minima). At all sites, both SAE and g show a strong variation with aerosol particle loading. The lowest values of g are always observed together with low σsp values, indicating a larger contribution from particles in the smaller accumulation mode. During periods of high σsp values, the variation of g is less pronounced whereas the SAE increases or decreases, suggesting changes mostly in the coarse aerosol particle mode rather than in the fine mode. Statistically significant decreasing trends of σsp are observed at 5 out of the 13 stations included in the trend analyses. The total reductions of σsp are consistent with those reported for PM2.5 and PM10 mass concentrations over similar periods across Europe."

2) The results sections in Chapter 3 require a substructure to become more readable. Currently, paragraphs are too long and there is no line of arguments the reader can follow. Instead the paragraphs are highly descriptive and do not point at the key messages of the data analyses. In its current form, the reader likely misses a large part of the information contained in this manuscript.

Following the Reviewer comment, Section 3 has been changed in the revised version of the manuscript. The presentation of the results is also different. In the revised version of the manuscript we present the discussion from cleaner sites to polluted sites. In the new figures 2, 4, and 5, 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. All figures related to the two non-European stations of TRL and ZEP were moved in the supporting material.
This new way to present the results made the manuscript more readable. Moreover, new paragraphs in Section 3 were added.

Section 3 was reorganized and the following sub-sections were introduced:

3.1.1 σ_{sp} at Arctic/Antarctic observatories;
3.1.2 σ_{sp} at mountain observatories;
3.1.3 σ_{sp} at coastal observatories;
3.1.4 σ_{sp} at regional/rural observatories;
3.1.4 σ_{sp} at urban/suburban observatories

3.2.1 Variability of SAE by geographical sector;
3.2.1 Variability of SAE by station type.

3.3.1 Variability of *g* by geographical sector
3.3.2 Variability of *g* by station type

Moreover, in the revised version of the manuscript the Section 3.4 presents the seasonal cycles, whereas the Section 3.5 presents the SAE and *g* vs. σsp relationships.
Section 3.4 was divided in different subsection in order to present the results from cleaner environments to polluted environments:

3.4.1 Seasonal variability at Arctic observatories;
3.4.2 Seasonal variability at mountain observatories;
3.4.3 Seasonal variability at coastal observatories;
3.4.4 Seasonal variability at regional/rural observatories;
3.4.5 Seasonal variability at urban/sub-urban observatories.

3) The conclusions section is not well structured and a lot of information may get lost. Sharpening and shortening of this section is recommended. Finally, what are the key

points of the presented work? This should be clearly expressed.

Following the Reviewer comment, the conclusions were modified in order to highlight the most important results from this study. However, given the large amount of information provided by this work, it was difficult to further shorten the Conclusion section. The modified Conclusion section is reported below:

"**Conclusions**

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

- An increasing gradient of σ_{sp} is observed when moving from remote environments (Arctic/mountain) to regional and to urban environments. At regional level in Europe, σ_{sp} also increases when moving from Nordic and Baltic countries and Western Europe to Central/Eastern Europe whereas no clear spatial gradient is observed for other station environments. For example, the lack of a clear spatial gradient of σ_{sp} measured at mountain observatories is likely due to the different altitudes of the observatories in the different geographical sectors considered in this study. Among the European mountain observatories a relationship was observed between station altitude and the median σ_{sp}, this latter being the highest at the station located at the lower altitude and vice versa.
- Overall, the highest σ_{sp} values are measured at low altitude observatories in Central and Eastern Europe and at some urban observatory in Southern Europe whereas the lowest σ_{sp} values are observed at mountain stations and at Arctic and Antarctic observatories. Low σ_{sp} levels, comparable to those measured at mountain sites, are also observed at the majority of the regional Nordic and Baltic observatories. The σ_{sp} values in Western Europe are on average higher compared to those measured in the Nordic and Baltic regions and lower compared to those measured at a regional level in Southern Europe. Some exceptions to these general features are however observed.
- The SAE does not show any clear gradient as a function of the placement of the station. However, a West to East increasing gradient is observed for both regional and mountain placements suggesting a lower fraction of fine-mode particle in Western/Southwestern Europe compared to Central and Eastern Europe where the fine-mode particles dominate the scattering.
- In fact, in Central and Eastern Europe, independently of the station placement, the SAE is among the highest observed across the network, indicating a large predominance of fine particles. In these regions, the SAE is even higher in summer compared to winter, suggesting the shift toward the small end of the aerosol particle size distribution likely linked to new particle formation events during the warmest months. On average SAE is lower in the Nordic and Baltic and western geographical sectors (likely due to the contribution from coarse-mode sea salt particles), and southern sectors (likely because of the presence of mineral dust particles from African deserts), compared to Central and Eastern Europe.
- The *g* does not show any clear gradient by station placement or geographical location reflecting the complex relationship of this parameter with the aerosol particles properties such as size distribution, particle shape or refractive index.
- Slightly higher *g* values are observed in Western Europe compared to Central and Eastern Europe. These differences in the *g* values, even if small, are consistent with the opposite gradient observed for SAE, this latter being smaller in Western Europe. However, the station-averaged *g* in Central and Eastern Europe is similar to the mean *g* observed in the Nordic and Baltic regions and in Southwestern Europe. Thus, contrary to the SAE, a clear relationship between aerosol size and *g* is not observed.
- Seasonal cycles for σ_{sp}, SAE and *g* are observed in all geographical sectors and explained by different factors. The seasonal cycles are especially marked at a regional level in Central and Eastern Europe where winter time episodes linked with stable air and thermal inversions favour the accumulation of pollutants. In these European regions the SAE (*g*) is higher (lower) in summer compared to winter due to variations in particle number size distribution due to the enhanced formation of small and optically active particles during new particles formation and subsequent growth. Clear annual cycles are also observed at mountain sites where σ_{sp} is higher in summer because of the enhanced influence of the boundary layer. In some cases, the SAE (*g*) is also high (low) in summer at mountain sites indicating a higher PBL anthropogenic influence during the warmer months and/or new particles formation episodes. In the Nordic and Baltic regions, the seasonal variation of σ_{sp} is less pronounced compared to Central and Eastern Europe, likely due to the different

meteorology and less pronounced PBL variations. Despite the relatively small σ_{sp} seasonal cycles in the Nordic and Baltic regions, SAE (g) increases (decreases) in these regions in summer compared to the winter period likely due to a season-dependent transport of air masses at these remote sites and the enhanced formation of secondary organic aerosols previously observed at these sites during the warmest months. At coastal sites in Northwestern Europe, the presence of sea-salt particles in winter also contributes to the observed pronounced seasonal cycles of SAE and *g*. in Southern Europe the seasonal cycles are strongly driven by the enhanced formation of secondary sulphate and organic matter in the summer, together with frequent Saharan mineral dust outbreaks.

- The analyses of the systematic variabilities of SAE and *g* as a function aerosol loading (σ_{sp}) reveal some common patterns. At all stations, *g* shows the lowest values at very low σ_{sp} likely because of the formation of new particles in a clean atmosphere followed by condensation/coagulation with, as a consequence, the generation of small but optically active particles. The *g* value then sharply increases with increasing □sp, indicating the shift of the particle number size distribution toward the larger end of the accumulation mode. Then, during periods of high σ_{sp} values, the variation of g is less pronounced at the majority of the stations, contrary to the SAE, which increases or decreases, suggesting changes mostly in the coarse aerosol particle mode rather than in the fine mode. At the majority of Northwestern, Central and Eastern European stations, the SAE maintains high values at high σ_{sp} values, indicating that the high σ_{sp} is dominated by fine particles. Conversely, at some sites in Southern Europe the SAE reaches values of around one or lower for high particle loads, indicating that, at these stations, the high σ_{sp} is dominated by mineral dust coarse particles mainly from African deserts. Exceptions are two urban sites in Southwestern Europe where fine particles, probably generated for the most part by traffic (and also from biomass burning) on average dominate the highest measured σ_{sp} values.

- The analyses of the trends reported in this investigation provide evidence that both extensive and intensive aerosol optical properties have significantly changed at some of the locations included here over the last 10 and 15 years. The σ_{sp} decreasing trends reported here are statistically significant at 5 out of 13 stations included in the analyses. These 5 stations are located in the Nordic and Baltic regions, and the central and southwestern sectors. Conversely, σ_{sp} trends which are decreasing are not statistically significant in Western and Eastern Europe. Statistically significant decreasing trends of SAE are observed at 3 out of 10 observatories included in the analysis: one site in the Nordic and Baltic sector and two mountain sites in the western and eastern sectors. These negative trends could be ascribed to reduced fine-mode anthropogenic emissions, as already observed in the literature for columnar SAE in Europe. Conversely, at two stations (one mountain site in Central Europe and one urban site in Southwestern Europe), the SAE shows a statistically significant increasing trend, suggesting a shift in the accumulation-mode particles toward smaller sizes and/or a change in the coarse aerosol mode. At the remaining 5 observatories, the reported SAE trends are not statistically significant. The backscatter fraction shows a statistically significant increasing trend at 5 out of the 9 sites where BF measurements are available. At three stations (the mountain site in Central Europe, the urban site in Southwestern Europe and one of the two sites in the Nordic and Baltic sector), both BF and SAE increase, suggesting consistent evidence of a shift in the accumulation-mode particles toward a smaller size. Conversely, at the other site in the Nordic and Baltic sector and at one mountain site in the western sector the BF increases whereas the SAE decreases.
- A general agreement is observed between the trend analyses preformed in this work and the analyses presented in a previous work confirming the general decreasing trends observed for σ_{sp} in Europe. However, some differences are also observed and likely due to the relative short periods used in these trend analyses and the different sensitivity of the methods used to missing values or presence of outliers. (Mann-Kendall or Theil-Sen vs. GLS/ARB or MLS; means vs. medians; different time granularity)

In conclusion, this investigation provides a clear and useful picture of the spatial and temporal variability of the surface in situ aerosol particle optical properties in Europe. The results presented here give a comprehensive view of the particle optical properties and provide a reliable analysis of aerosol optical parameters for model constraints. In addition, the analyses presented here suggest findings that may need additional investigation. For example, the fact that at some of the stations the trend of □sp changes in terms of both statistical significance and sign depending on the period used, suggests that trend analyses are necessary in the future when longer-duration records will be available. Moreover, the fact that at some sites BF and SAE show different signs in their trends suggests that further analysis is needed to better understand how other aerosol parameters, such as particle size distribution and mean diameter, affect the relationships between BF and SAE."

4) ***Consideration of published results on the long-term evolution of aerosol optical properties: Although previous studies on aerosol scattering properties (see, e.g., Andrews et al. (2011), Collaud Coen et al., (2013) and Zanatta et al., (2016)) are***

mentioned in the text, they have been included into the discussion only briefly. A discussion is missing (or got lost due to the current organization of the manuscript) whether the results presented here are in agreement with the published results, or whether they provide new findings, and then the question would be: where do differences come from? Finally, the discussion of the evolution of aerosol light scattering properties together with light absorption properties published in Zanatta et al. (2016) is completely missing.

The comparison with Zanatta et al. (2016) is presented in the revised version of the manuscript. To compare with absorption measurements from Zanatta et al. (2016) the following sentence was added to the paragraph 3.1:

"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 σsp 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 σsp. 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."

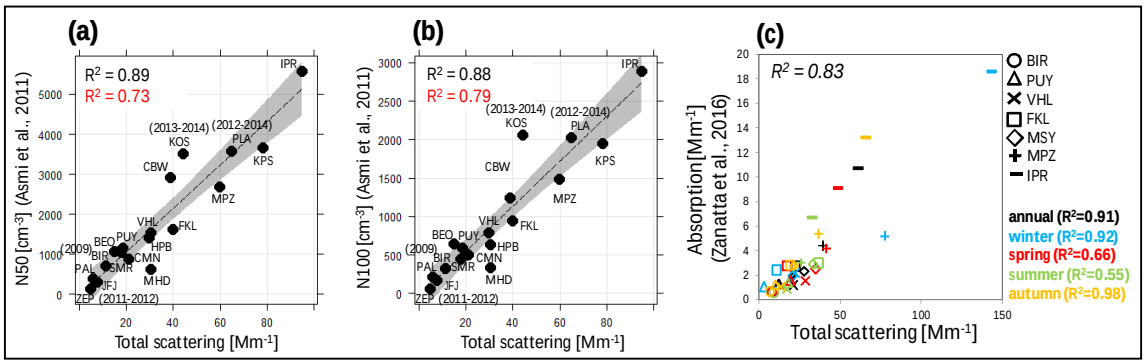

Figure 3: Relationship between: (a) N50 (mean particle number concentration between 50 nm and 500 nm), (b) N100 (mean particle number concentration between 100 nm and 500 nm), (c) absorption coefficient and mean aerosol particle total scattering coefficient. (a) and (b): data averaged over the period 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).

Comparison with the trend analysis performed by Collaud Coen et al. is reported in Section 3.6.3. In this Section we also provide possible reasons for the small discrepancies observed compared to the work from Collaud Coen et al. The sentence is reported below:

"These differences are thus likely due to the relative short period used in these trend analyses and the different sensitivity of the methods used to the presence of missing values or outliers especially at PAL where σsp is very low (cf. Fig. 2). For example, in this work the SAE calculated for PAL during the year 2007 was removed from the trend analysis due to the presence of too many extreme high SAE values, thus also likely explaining the difference observed for SAE with the work from Collaud Coen et al. (2013). Moreover, here we use de-seasonalized monthly means for trend analyses whereas Collaud-Coen et al. (2013) used de-seasonalized medians with different time granularity (3 days) thus likely affecting the comparison, especially over relatively short periods."

Moreover, the results of the comparison of trend analyses with the previous work from COllaud Coen et al. is also highlighted in the Conclusion section in the revised version of the manuscript.

Comparisons with the work from Andrews et al. (2011) are reported throughout the text. Below the sentences:

Variability of SAE:
"This high variability of SAE at mountain sites was also reported by Andrews et al. (2011). Andrews et al. (2011) reported SAE values from 11 mountaintop stations worldwide ranging from less than one to more than two."

Seasonal cycles at mountain sites:
"Similar findings were for example already reported by Nyeki et al. (1998) for JFJ and summarized by Andrews et al. (2011) for many mountain top stations worldwide and by Pandolfi et al. (2014) for MSA station."

g-σsp relationship:

"The asymmetry parameter g shows the lowest values under very low σsp suggesting the predominance of small fine mode particles. Andrews et al. (2011) reported similar g-σsp relationships at different mountain sites and suggested that the removal of large particles by cloud scavenging or by deposition during transport could explain the observed low g under a clean atmosphere. They also suggested that the formation of new particles followed by condensation/coagulation could generate small but optically active particles. Here, we show that this behavior of BF or g as a function of σsp was observed at all sites, not only at mountain sites. "

"Andrews et al. (2011), Pandolfi et al. (2014) and Sherman et al. (2015) showed that BF tends to decrease with increasing aerosol loading, consistent with the observed increase of g."

SAE-σsp relationship
"Andrews et al. (2011), Pandolfi et al. (2014) and Sherman et al. (2015) showed that BF tends to decrease with increasing aerosol loading, consistent with the observed increase of g."

Variability of g by station type:
"On average, g values range between 0.49 to 0.64 at mountain sites with a mean value of 0.58±0.05. This value is consistent with the mean value of 0.61±0.05 reported by Andrews et al. (2011) at the mountain sites included in their work."

MINOR COMMENTS

5) ***In section 2.2.2.1, the applied truncation correction is described. For light absorbing aerosol (single scattering albedo < 0.8) a method proposed by Müller et al. (2011) is applied. However, Massoli et al. (2009) presented a correction scheme particularly for light-absorbing aerosols measured with the TSI Model 3563 Integrating nephelometer. It should be briefly discussed why this approach has not been applied at the stations running TSI Model 3563 instruments.***

It is important to clarify that at all sites the correction for non ideal illumination and truncation are performed using the Angstrom based approach provided by Muller et al. (2011) for the Ecotech AURORA3000 and by Anderson and Ogren (1998) for the TSI nephelometer. Thus, no other methods for

correction are used by data providers. This is mostly due to the simplicity of the Angstrom based methods provided by the aforementioned papers. We think that the analysis of the different sensitivity of different correction schemes to different concentration of light absorbing species is far behind the objective of this work.

To clarify this point, the beginning of the section 2.2.2.1 has been 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 0-10º) and near-backward (around 170-180º) 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)."

6) In section 2.2.2.3, wavelength adjustments were conducted for sites where multiplewavelengths data do not exist. For that purpose SAE values were prescribed (1.5 for FKL and SIR and 2.0 for CMN). The choice of these SAE values should be justified.

Given that the SAE is not available at SIR and FKL, in the revised version of the manuscript we calculated the differences after λ-adjustment using a reasonable range of SAE values (1.0 to 1.5). For CMN the SAE = 2 was calculated using data from the 3-λ nephelometer available from 2014 at CMN (before 2014 the 1-λ nephelometer was used)."

The section 2.2.2.3 was modified as follow:

"2.2.2.3 Available wavelengths
In this work we present and discuss the σsp, backscatter fraction (BF) and asymmetry parameter (g) measurements obtained using the green wavelength of the integrating nephelometers. The available wavelengths ranged from 520 nm (2 stations; CMN and VHL) to 550 nm (18 stations). Other wavelengths used are 525 nm (6 stations) and 532 nm (used at FKL until 2010; cf. Table 2). An exception is SIR, where only σsp values at 450 nm are available. The measurements of σsp reported here are not adjusted to 550 nm, which is generally the most common wavelength (e.g. Andrews et al., 2011) because of the different data availability of σsp and SAE at the measuring stations. As discussed in the following sections, the SAE is calculated for σsp data higher than 0.8 Mm-1, thus leading to different data coverage for σsp and SAE and preventing the adjustment of all measured σsp to 550 nm. Moreover, the SAE is not available at FKL and SIR (or at CMN until 2014) thus preventing any wavelength adjustment at these stations. Using the mean SAE calculated at those stations, where σsp is measured at wavelengths in addition to 550 nm (cf. Tables S4 and S5 in Supporting material), we estimate differences in the sp values of less than 6% after adjusting to 550 nm. 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. Finally, at CMN, the effect of the adjustment of σsp to 550 nm (from 520 nm) using a mean SAE of 2 (calculated using the 3-λ nephelometer data from 2014; cf. Table S5) is below 10%."

7) Before resubmission, checking of the language by a native speaker is highly recommended.

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