

We thank the Reviewer#1 for his/her comments.

We added the following sentence to the Acknowledgments section to acknowledge the detailed and helpful comments to the manuscript from the two anonymous Referees and the co-editor Andreas Petzold.:

“We also thank the co-editor Andreas Petzold and the two anonymous reviewers for their constructive comments.”

Below the answers to the Referee’s comments. The modified text is highlighted with the yellow color in the 2d revision of this manuscript.

The authors have done a great job of responding to almost all of my comments. The paper flows much better! I just have a few minor things that I think should still be addressed or at least considered.

**1) Response #1 about comparing with Zanatta**

That’s a great addition. Lines of constant SSA could be added to the plot. I know you mentioned (response#2) that someone else is writing an SSA paper, but adding those lines shouldn’t interfere with that SSA project.

The sentence describing Figure 3 was modified as it follows:

“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  $\sigma_{sp}$  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  $\sigma_{sp}$ . 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 3c also reports the mean single scattering albedo (SSA). Figure 3c also reports the mean single scattering albedo (SSA). On average lower SSA is observed at IPR, whereas higher SSA is observed at the Nordic and Baltic VHL and BIR observatories.”

Figure 3 (reported below) was modified accordingly to the Reviewer comment.

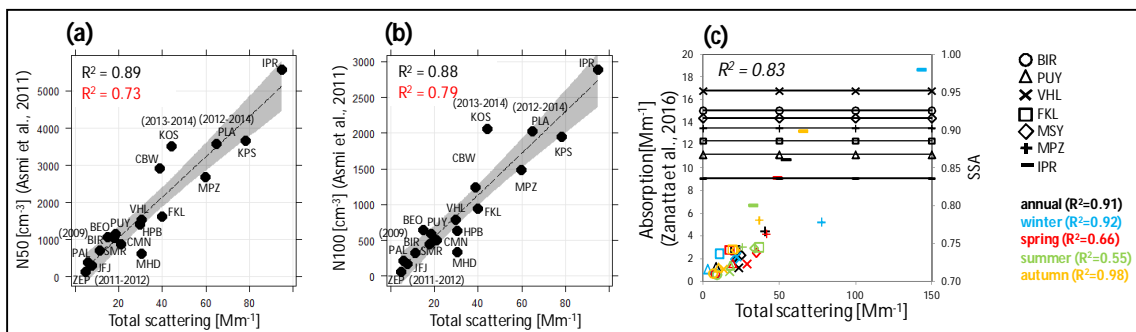


Figure 3: Relationship between: (a) N50 (mean particle number concentration between 50 nm and 500 nm), (b) N100 (mean particle number concentration between 100 nm and 500 nm), (c) absorption coefficient and mean aerosol particle total scattering coefficient. (a) and (b): data averaged over the period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

**2) Response #13 about skewness**

The authors added some more text about skewness but it doesn’t actually get at what I was asking about. ? I was hoping to see something more in the context of aerosols and less

**mathematical. With the new text I still don't think it's really clear why a reader should care about skewness or what the implications are for aerosols. For example, what does skewness say about extreme events (if anything)? Or aerosol variability? Etc...**

To properly answer the Reviewer comment 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 ssp for each station, evidencing the presence of these right-skewed tails."

Was replaced with the following sentence:

"The skewness can be used to evaluate the asymmetry of a distribution. 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. For example Querol et al. (2009) used the skewness to assess the importance of Saharan dust outbreaks on PM<sub>10</sub> levels measured at different sites across the Mediterranean basin. They found a positive correlation between the calculated skewness and the net dust contribution to the measured PM<sub>10</sub> concentration (i.e. the strength of dust pollution episodes; cf. Fig. 6 in Querol et al., 2009). Figure S2 in the Supporting Material shows the frequency and cumulative frequency distributions for  $\sigma_{\text{ssp}}$  for each station, evidencing the presence of these right-skewed tails. "

Querol, X., Pey, J., Pandolfi, M., Alastuey, A., Cusack, M., Pérez, N., Moreno, T., Viana, M., Mihalopoulos, N., Kallos, G., Kleanthous, S.: African dust contributions to mean ambient PM<sub>10</sub> mass-levels across the Mediterranean Basin, *Atm. Env.*, 43, 4266–4277, doi:10.1016/j.atmosenv.2009.06.013, 2009.

### **3) Response #15 about cut sizes**

**The authors did a bunch of work to look at this question of whether inlet cut size changes affected the resulting aerosol climatology at the sites they looked at. I don't think the figures they generated to respond to this question need to be added to the manuscript or supplemental materials but I think it would be useful to add one or two sentences to the manuscript to reflect this additional work they did. For example, right now there's a sentence that says "Consequently, the inlet change from PM1 to PM10 at KPS had probably only a minor effect on SAE." The authors have now looked at the data and can say something a little more definitive than 'probably'.**

**Also, there should be some mention of size cut changes in the methods section. I think the way to do this is to make an additional methods section: 2.2.2.4 'Inlet size cut changes'. There the authors should note that inlet size cut change could potentially affect both scattering and SAE and trends. I would move the parts of the second paragraph of section 3.2 describing the inlet size cut changes and previous investigations on the effects of size cut changes to this new section. Then I would suggest adding a few sentences to section 2.2.2.4 saying that the authors looked at climatologies of SAE (and scattering?) for different inlet sizes and for different time periods for and didn't see any obvious changes in the climatology as a function of size cut due to interannual variability (although I'm not sure that's totally true for KPS!). Then in section 3.2 you can just refer to section 2.2.2.4. Also in section 3.6 where you mention size cut issues you can again refer back to section 2.2.2.4.**

Following the Reviewer suggestion a new section (2.2.2.4) was added to the manuscript. All parts of the manuscript referring to the inlet changes were moved to this new section. The new section is reported below:

#### “2.2.2.4 Inlet size cut changes

It should be noted that any comparison of the  $\sigma_{sp}$  and SAE values among the different stations and the presented trend analyses could be slightly biased by the different particle size cuts upstream of the integrating nephelometers used in this work (cf. Table 1). Currently, all ACTRIS integrating nephelometers measure whole air or PM10, with the exception of SIR, where the PM1 inlet is used. Whole air is currently measured at mountain observatories (BEO, CMN, JFJ, PUY, CHC), one coastal observatory (MHD) and one urban observatory (UGR) (cf. Table 1).

At some stations, the inlet was changed from whole air to PM10 at some point, namely at OPE, FKL and TRL. Given the lower scattering efficiency of aerosol particles larger than 10  $\mu\text{m}$ , no important differences in the aerosol particle optical parameters should be expected between aerosol particles sampled with a whole air and a PM10 cut-off. At the other stations the inlet was changed during the measurement period from a cut-off lower than 10  $\mu\text{m}$  (1  $\mu\text{m}$  at KPS; 2.5  $\mu\text{m}$  or 5  $\mu\text{m}$  at PAL, MSA and MAD) to PM10. For PAL (where a median SAE of around 1.8 was measured; cf. Paragraph 3.2 and Table S5), Lihavainen et al. (2015a) assumed that the inlet changes (from PM5 to PM2.5 in 2005 and from PM2.5 to PM10, cf. Table 1) had only minor effects on scattering because the number concentration of coarse particles is very low at PAL. Similarly, the KPS observatory registers among the highest SAE values observed in the network (median value of around 2; cf. Paragraph 3.2 and Table S5) suggesting an aerosol particle size distribution dominated by fine particles. Moreover, at KPS, the inlet was changed in April 2008, less than 1.5 years after the measurements commenced, and thus likely has also a minor effect in the trend analyses and climatology performed at this site over the period 2006 to 2014. Two stations (MSA and MAD) changed the inlet from a PM2.5 diameter cut-off to PM10. For these two Southern European stations the inlet change may have had an effect on the SAE, especially during Saharan dust outbreaks, which are however sporadic events. Finally, the FKL observatory was removed from the trend analysis because the inlet was changed from whole air to PM10 in 2009, from PM10 to PM1 in 2011 and again from PM1 to PM10 in 2013 (cf. Table 1). These events likely had a major effect on the measured particle optical properties.

A sensitivity study (not shown) was performed to assess the effect of the inlet changes on the SAE values measured at the aforementioned stations. We looked at the climatology of SAE for different inlet sizes and for different time periods (with and without inlet size changes) and we did not observe any obvious change in the climatology as a function of size cut due to interannual variability. Thus, despite the differences in the particle diameter cut-off, the comparison between the different stations seems feasible.”

**Note: the manuscript says that MSA and MAD changed from PM2.5 to PM10 but the plot made for MSA in the response to the reviewers for comment#15 only shows box whiskers for changes between whole air and PM10. Not sure if that's a typo.**

We are sorry for this. It was a typo. MSA and MAD changed from PM2.5 to PM10.

#### 4) Section 3.1.3

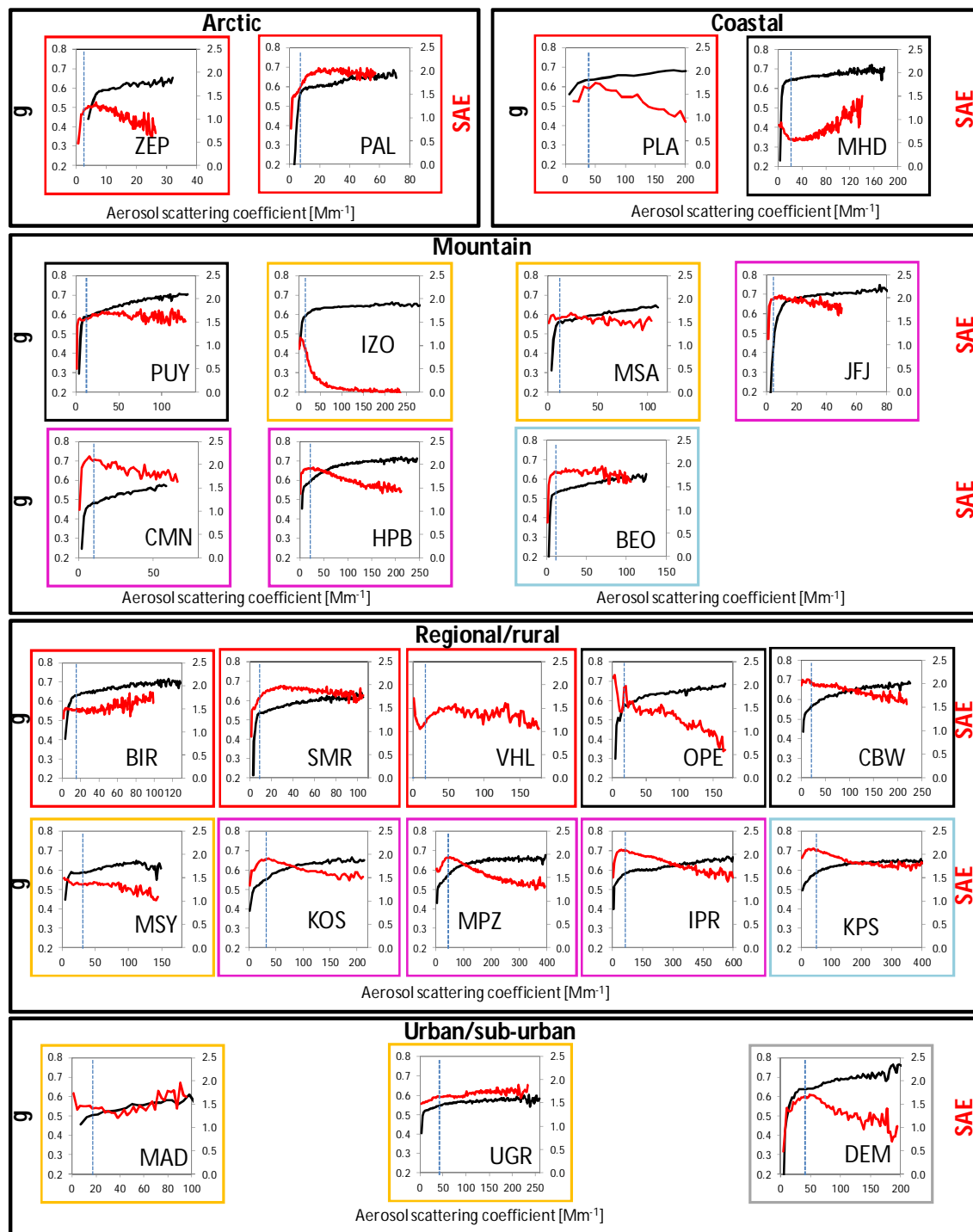
**Last sentence refers to 'Paragraph 3.5' – do the authors mean section 3.5?**

Yes, we mean section. The text was accordingly modified.

#### 5) Figure 9

**The discussion about Figure 9 in Section 3.5 would probably be easier to follow if the y-axes for g and SAE were the same for all plots.**

The Figure 9 was changed accordingly to the Reviewer comment. The new Figure 9 is reported below.

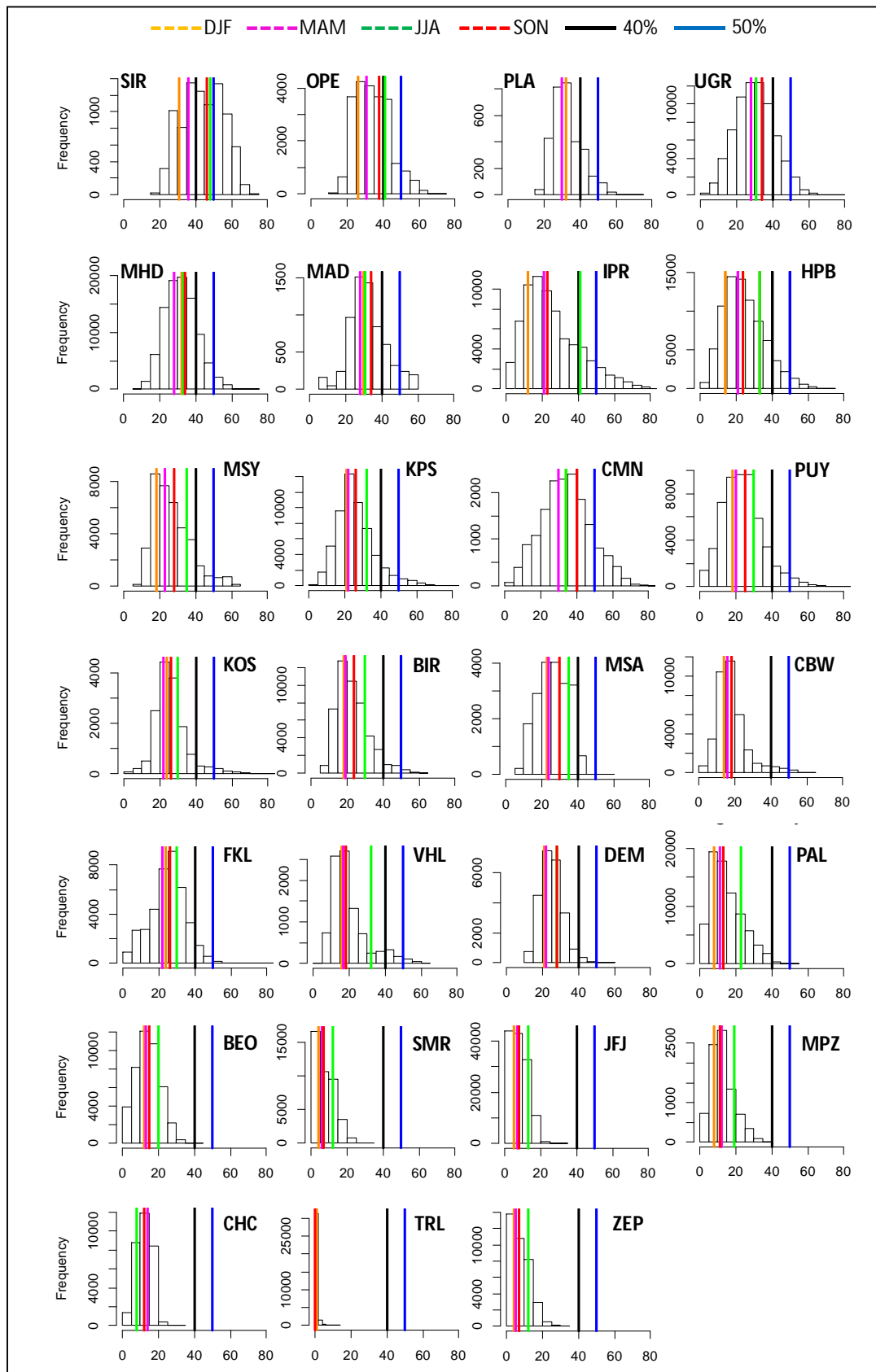


**Figure 9:** Scatterplots between  $\sigma_{sp}$  (x-axes) and SAE (right y-axes; red lines) and  $g$  (left y-axes; black lines). Dashed lines represent median  $\sigma_{sp}$  values at each station. Different colours highlight different geographical locations as in Figures 2, 4 and 5.

**6) Figure S1**

The dashed vertical lines are really hard to see. Could you make them solid like the other two lines?

The vertical lines in Figure S1 were changed accordingly to the Reviewer comment. The new Figure S1 is reported below.



**Figure S1:** Frequency distributions of sampled RH at ACTRIS observatories. Vertical lines: 40% GAW recommendation (black); 50% RH threshold used in this work (blue); median RH in winter (yellow); median RH in spring (magenta); median RH in summer (green); median RH in autumn (red).