

### **Answers to anonymous referee 3:**

The authors thank the referee for their detailed review of the manuscript and for all their comments and suggestions allowing a clear improvement of the paper.

During the review process, the routines for MK trend analysis were translated into R and an error was found in the selection of data for north hemispheric winter season. This error was corrected in the original matlab routines leading to minor changes in slope absolute values for most of the stations, but also sometimes to modification of the statistical significance. The more important changes are:

- ALT was the only station with ss trend in absorption coefficient and this was the only case where there is a strong discrepancy among the analysing methods, MK being ss positive, LMS/log not ss and GLS/day ss negative. The correction leads to MK not ss trend in absorption coefficient at ALT and remove therefore the solely strong discrepancy between the methods.
- MLO has a ss negative trend in scattering coefficient for the last 10 y, leading to a better agreement between scattering and absorption trends. The evolution from positive to negative ss trends is now well established.
- Some other not ss present-day trends are now ss negative (RMN scattering coefficient, CPR absorption coefficient, THD single scattering albedo) or ss positive (PUY single scattering albedo, MSY scattering Ångström exponent, LLN absorption Ångström exponent).
- Some ss trends are now not ss: IZO absorption coefficient,
- One trend (JFJ scattering Ångström exponent for the 20y period) change from ss negative to ss positive trend.
- The statistical significance of some of the 10 y trends of the time evolution analysis (Sect. 3.2) is also modified, but these changes do not impact the results.

The revised manuscript and all tables and figures were corrected in order to take into account the new results.

### **Answers to specific comments:**

1. P3, L10 and L11: there is a space between - and 0.45 (leading to a newline between)

*Thank you, the space was removed.*

2. P8, L15: Assuming an Absorption Angström exponent of one for SSA calculation could cause further dependence on changes of size distribution or chemical composition. What is the impact of this assumption?

*Yes, this assumption can lead to a SSA departing from the true values. Let consider a range of absorption Ångström exponents between -0.5 and -2 and an often encountered ratio between scattering and absorption value of 10. An adjustment from blue (470 nm) to*

green wavelength (570 nm) would lead to an error of 10% and -18% for  $\hat{a}_{ap} = -0.5$  and  $-2$ , respectively. An adjustment from red (660 nm) to green wavelength (570 nm) would lead to an error of -7% and 16% for  $\hat{a}_{ap} = -0.5$  and  $-2$ , respectively. This will induce a maximum error of  $\pm 1.6$  % on the SSA values. A similar difference for the scattering Ångström exponent would lead to a maximum error of  $\pm 2$  % on the SSA values considering the TSI wavelengths (450, 550 and 700 nm). A combination of maximal error on both  $\hat{a}_{ap}$  and  $\hat{a}_{sp}$  leads to a maximum cumulative error of 6% on SSA. Considering the large errors usually estimated to approximately 30% of the absorption coefficient and of 10-20% on the scattering exponent, the error induced by the  $1/\lambda$  dependence can be considered as negligible.

3. P8, L24 – L28: is part of data preparation and thus could be moved to section 2.4

*This was done in the revised version.*

4. Section 2.4 is missing a paragraph on assessment for nephelometer artefacts

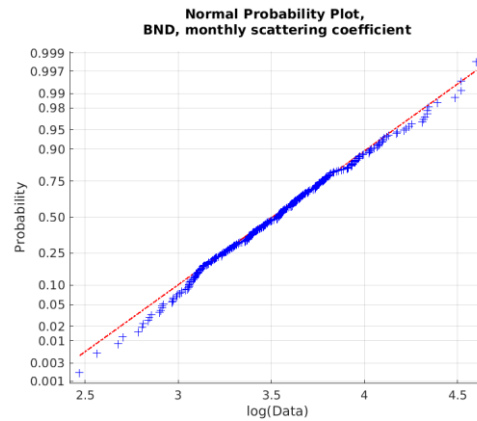
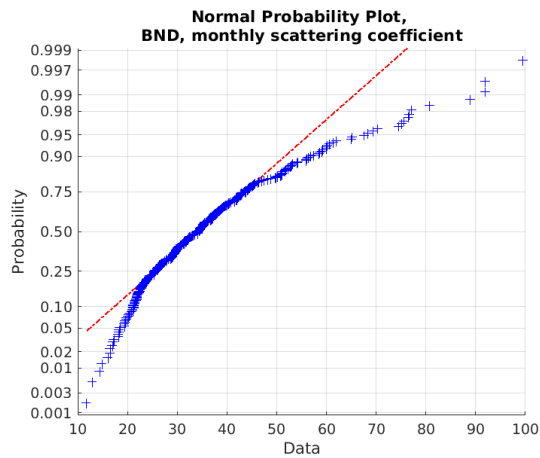
*The truncation error and the ways it is considered for the various instruments used in this study are described in sect. 2.2, second §. The artifacts bounded to the humidity percentage during the measurement are described in sect. 2.2, third §, in sect. 2.4 (p. 9 lines 45) as well as discussed in sect. 4.1 (p. 21, line 37). The way to handle the wavelength dependence, including the computed parameters, is described in sect. 2.3. A new § was added to Sect. 2.4 in order to describe the potential effects of the truncation correction on the trend analysis:*

*“4) Nephelometer truncation correction artefacts: as explained in Sect. 2.2, the various types of nephelometer measure at different truncated angular ranges that were corrected by several algorithms or even not corrected. The absence of truncation correction leads to lower scattering and backscattering coefficients than the true values and the correction algorithm effects are known to increase with particle size. The most important requirement that was verified for this trend analysis is the coherent treatment of nephelometer data for each time series. The bias leading to a higher contribution of Aitken and accumulation modes than the coarse mode is difficult to estimate, but the minimal differences in PM1 and PM10 results (see Sect 4.2) suggest this artefact is small. The effect of the humidity on the nephelometer measurements is regarded as the most significant artefact.”*

5. P12, L15: do the monthly medians fit the log-normal distribution and what was the procedure to deal with negatives or zero values? What was the reason for median as aggregation method?

*The monthly median can be considered (at least for part of the time series) as lognormally distributed (see normal probability plot thereafter). None of the values (e.g. negatives, zeros, very low values) were removed before computing the monthly medians. The monthly median aggregation leads to very few negatives that were discarded before taking the logarithm of the data. Aerosol time series do not have zeros. Absorption, scattering and backscattering coefficients have very low values that could be considered as below detection limit values, but no peculiar treatment was applied to very low values. Since most of the parameters analyzed in this study are not normally distributed, the median was chosen to minimize the effect of extreme values on the average (see sect.*

2.3 first §). This is the usually recommended method for aggregation in case of not-normally distribution.



6. P16, L13: could add “Backscatter fraction (b)” for readability

*This was done in the revised version.*

7. P20, L15 and L17: “derived parameters” would be more specific (instead of “computed parameters”)

*This was modified in the revised version.*

8. P23, L18 and L19: the intention of “Ideally, abatement policy...” is not clear and vague

*The abatement policy mentioned here concern the governmental regulation to decrease atmospheric pollutants and comprise both gaseous and particle emissions. The manuscript was modified to clarify this point: “Ideally, abatement policy aimed at decreasing atmospheric pollutant levels would take into account both climate and health impacts.”*

9. P27, L15: “due because”

*Thanks, “due” was removed.*