

We would like to thank Referee #1 for their very positive feedback on the paper and their comments, which are addressed point by point below. Note that in addition to the marked edits, the paper has undergone English editing resulting in minor changes that are not systematically highlighted in the revised manuscript.

Comment 1: Page 5, lines 10: There have been many long-term trend analyses of optical properties, number concentration, and PNSD. Does this refer to analyses involving several sites? Please clarify in the text.

Reply 1: If the Reviewer refers to the studies by Asmi et al. (2013) and Collaud Coen et al. (2013), they in fact involve several sites. This is now explicitly mentioned in the text:

“Moreover, Asmi et al. (2011) reported on the variability of the PNSD, also in Europe, based on measurements collected at 24 sites; shortly after, the first multi-site long-term trend analyses of aerosol optical properties (Collaud Coen et al., 2013), number concentration and PNSD (Asmi et al., 2013) were performed using measurements conducted at stations located in Europe, North America, Antarctica and on Pacific Ocean islands.”

Comment 2: Page 6, line 10: Why 2016 and 2017? Please explain in the text.

Reply 2: The present study is focussed on 2016 and 2017 for consistency with the analysis of Laj et al. (2020), which our work aims to complement. These specific years were selected as they provided the most updated view of measurements worldwide when the study of Laj et al. (2020) was initiated. The text was modified as follows:

“Data collected at 62 sites contributing to SARGAN in 2017 or 2016, i.e. the reference years as chosen in Laj et al. (2020) (see more details about data availability and coverage criteria in Sect. 4), were included in the present work, among which 57 were already involved in the short analysis of the total number concentration reported in that study.”

Comment 3: Figure 1 and throughout: The symbols in the legend box should be larger.

Reply 3: The size of the symbols has been increased in the legends. The triangles in figures 3, S3 and S4 have also been slightly enlarged to ease the distinction between downward and upfacing triangles.

Comment 4: Figure 1 caption: It would be helpful if MPSS were spelled out.

Reply 4: The last sentence of the figure caption was changed to account for this suggestion:

“The sites operating a mobility particle size spectrometer (MPSS) are additionally marked in italic bold.”

Comment 5: Page 6, line 18: What is meant by “(sub)-urban”? Does it include both suburban and urban? As far as I can tell neither it or “suburban” are used anywhere else in the paper.

Reply 5: In fact, the expression “(sub)-urban” was meant to refer to urban and suburban sites but, as they are all referred to as urban in the rest of the paper and also in Laj et al. (2020), this expression was removed to avoid confusion:

“The stations are classified based on the combination of a geographical [...] and footprint (rural background, forest, urban, pristine or mixed) criteria as introduced in Laj et al. (2020).”

Comment 6: Page 7, line 9: What is the definition of “large” particles as used here?

Reply 6: “large” refers here to particles that are above the measurement range of the instrument. The sentence was slightly change to clarify this point and two references were added:

“As part of this quality control process, negative concentrations arising from inversion issues in certain conditions (e.g. presence of particles above the size range covered by the MPSS, such as dust or sea salt; Pfeifer et al., 2014; Wiedensohler et al., 2018) were filtered out”.

Comment 7: Figure 10: It is difficult to tell the difference between the size of the symbols. Please change so that the size difference is clear. The yellow symbols are difficult to see. Can these be changed to another color?

Reply 7: To avoid any confusion, the different marker sizes have been replaced by different shapes (triangle and square), and yellow was changed to light orange to improve readability.

Please note that while adjusting the symbols, we realised that the size of the marker was not consistent between the two panels of Fig. 10. In Fig. 10.b, all correlations are in fact statistically significant at the 95% confidence level (instead of 90%); this has also been corrected in the text (P29, L25).

Comment 8: Also – up to here the coefficient of determination (r^2) has been used to describe the significance of the correlation between two parameters. Why is the Spearman's rank correlation coefficient being used here?

Reply 8: We used the coefficient of determination to evaluate the agreement between observed and fitted PNSD (i.e. same variable), following the same approach as Asmi et al. (2013). In the case of Fig. 10, as indicated P29, L4-5, the Spearman's rank correlation was used instead, similar to Collaud-Coen et al. (2018), as we evaluate the correlation between two different variables (particle concentration and elevation of the site or ABL TopoIndex) which are not necessarily normally distributed, and for which we cannot anticipate a linear relationship.

Comment 9: Page 40, line 5: change to “RESIDENTIAL”

Reply 9: Thank you for noticing the typo!