

GENERAL REMARKS

From the review by two referees and from my editor's review, the manuscript is still considered presenting high-relevance data and analyses of high interest for the research community. The authors have responded to the concerns raised by both referees, but still in an insufficient manner. I have sent the manuscript for a second review to one of the referees, who confirmed that many of the technical issues have been answered but the presentation quality is still considered poor.

Serious objections against publication arise still from the manner, the scientific results are presented. The core of the manuscript is the analysis of the long-term time series which is of high relevance. However, there is no red line to follow in this manuscript. The presentation of results oscillates between different foci which makes it hard to follow.

To help making the manuscript acceptable for publication, I suggest the following way forward:

- (1) The time-series of PM₁₀ data is well described but the reader has enormous difficulties identifying if the authors focus on dry or humid conditions. The confusion starts with the description of the sampling conditions in Section 2.2.1. Instead of combining all relevant information on sampling lines, aerosol drying etc. in this section the information is distributed among the Sections 2.2 and 2.3. This is in particular true for all topics related to relative humidity, humidity-driven particle growth and related impacts on aerosol optical properties.

Here, I suggest combining all information on particle sampling, including references to aerosol sampling at SMEAR, into Section 1. Already here it should be stated whether the manuscript is focusing on dry or ambient aerosol conditions. Having said this, the specific treatment of samples at high RH conditions can be added. But it should be clear to the reader if the authors generally focus on dry or ambient conditions. The treatment of humid cases can then be introduced as special cases. Obviously, there have difficulties caused by the failed humidity control of the sampling during certain period, but the description of the difficulties and the resulting effects on the data analysis are presented at different positions of the manuscript. This needs to be presented in one section to allow the reader a clear assessment of the quality of data and deduced results.

- (2) To which conditions (dry, humid) do the reported optical properties refer to? For instance, in Section 2.3, the authors start directly with the introduction of the humidity growth factor, but it is not explained why.

Here, a much clearer reasoning and description of the approach is needed. Having said this, it will be much clearer to follow the analyses. In particular, the discussion of the backscatter fraction needs to be removed at all since this factor was not corrected for humidity effects and thus treated completely different than the other optical properties. Thus, there is no way of comparing scattering and backscatter properties.

- (3) The authors state correctly that trend analyses of time series of less than one decade duration have to be taken with care; see Section 3.2 on page 14. Why do the authors then present in detail the trend analysis of the PM₁ time series? This is difficult to justify and contradicts with the statement made on trends from time series of less than 10 years duration.

Here, I suggest removing the PM1 data from the trend analysis but present them as comparison to the PM10 trend results. This would give less weight to the PM1 results in terms of trends but still allows showing the difference to the PM10 results.

- (4) There are intensive properties discussed with respect to trends, but the trends are not significant. These facts need to be reflected in the description of the results; see Table 3 for quantities α_{scat} , α_{abs} , and refractive index real and imaginary parts. The interpretation of results needs to be softened since the trends are not sufficient.
- (5) In the Figures there are clear statements missing to which fraction (PM10, PM1) the plots refer to. If the authors state at the very beginning on which fraction the manuscript is focusing at, these statements are no longer necessary.
- (6) In Tables 1 and 2, the authors present statistical analyses of aerosol optical properties. Inspecting the tables in detail shows that there are large discrepancies between average values and median values. Such discrepancies are always a clear sign that Gaussian statistics is not applicable. I suggest reducing the statistical analysis to the robust analysis of median and percentiles. By doing this, any biases caused by extreme values are avoided and the results are much more stable.

I am well aware that this further major revision may cause another large amount of work. On the other hand the paper has great potential and the trend analysis is of high interest for ACP but the presentation of the material requires a much clearer discussion of different topics and analysis results to improve overall readability and the delivery of a clear scientific goal/interest.

In summary, I encourage the authors to undertake this effort and accept the additional burden since the material is clearly worth it. I ensure further contribution and backing of the process to help publishing the manuscript. In case of questions or discussion the authors may contact me directly at a.petzold@fz-juelich.de.

Besides these general topics, I strongly encourage the consideration of the specific comments given by one referee in his 2nd review.

SPECIFIC COMMENTS BY REFEREE #2, arranged by section

1 INTRODUCTION

The section is incomplete and not well structured.

The importance of AOPs for the estimation of the RF is not mentioned. Thus, the climatic motivations at the base of this study are unclear. The aerosol-cloud interaction is, presently, not of primary need. The description of the measurements and site (P2L16-30) is not of necessary, since it can be extensively described in the method section. Generally, I also find a serious lack of references. An introduction on aerosol trends, such as previous works, environmental policies, dimming, and brightening might underline the importance of your work.

2 MEASUREMENTS AND METHODS

All measurements and data analysis are fully described. However, I would suggest to restructure and reorganize the different sections, starting from the titles. Try to be more specific and attract the interest of the reader avoiding general titles such as "The field site", which could be changed into a

more appealing “The SMEAR II boreal research station” (by the way here you have to insert the description given at P2L16-21). EFR needs a separate section. Sections 2.3.4 and 2.5 can be merged.

3.1 OVERVIEW OF THE DATA

Work on the titles and try to be a bit more original: “Characterisation of the boreal aerosol”, or “Scandinavian background aerosol optical properties”, etc... . It is important to define a target-topic for each section, I hardly see what you want to show here. The seasonal analysis can be used to describe the impact of different sources or the role of atmospheric processes on the AOPs.

3.2 TRENDS

A bit of rework on the red line and additional thinking on the climatic/environmental implications are needed.

3.3 AEROSOL OPTICAL PROPERTIES AND SIZE DISTRIBUTION

As already indicated by me and the second reviewer, I do not understand the goal of this section. I would definitely give more priority to the interpretation of trends rather than to the size distribution. Moreover, too many variables are discussed making the section quite chaotic. Potentially, a reduced/simplified discussion on the aerosol size distribution can be introduced in 3.1 as part of the aerosol characterization.

3.4 SEASONAL VARIATION

As said before, I would move and merge it with 3.1

3.5 VARIATION BETWEEN THE PM10 AND PM1 MEASUREMENTS

As already argued in the first review, this section is of scarce interest. In fact, a good part of the section is used to justify the differences from previous works (P20L27-P21L7) and the high uncertainty (P21L21-27).

3.6 RADIATIVE FORCING EFFICIENCY

The section reads nice and might represent the final outcome of the manuscript. However, I have some suggestions:

- a) The title can be improved: “Seasonality and trend of ”.
- b) The text is very intricated, limiting the understanding.
- c) Figure 7 tells that humidity is very important in summer (hence hygroscopicity is a critical property of the aerosol in order to asses RF) and that optical properties have a smaller impact of RF compared to environmental factors. Considering these two messages and the high number of assumptions for the RFE calculation, Fig. 8 is quite approximative. A full investigation of the impact of AOPs on RF would need a full separated paper. Thus, I would suggest excluding, this time, Fig. 8 and the subsequent (very short) discussion.

4 SUMMARY AND CONCLUSIONS

As a consequence of all the above comments, the conclusion section needs rethinking and rewriting.