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Interactive comment on "Over a ten-year record of aerosol optical properties at SMEAR II" *by* Krista Luoma et al.

Anonymous Referee #1

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GENERAL COMMENT

The here presented manuscript describes the variability of several aerosol optical properties (AOPs) measured in southern Finland for more than a decade. The multi-year variation of AOPs is presented together with a detailed analysis of AOPs variability on a shorter timescale. Due to its time coverage, the dataset presented here is of great relevance and might help to understand how aerosol changed in a North European back-ground site during the last ten years. The scheme and structure of the manuscript are linear and follows a logical order. However, the amplitude of the dataset generates a certain overloading of the manuscript, meaning that the results are not always properly discussed within a climatologic perspective but simply described. As a consequence, is difficult to identify the overall scientific message of the work. I truly

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believe that the paper covers the topic of interest of ACP, but I would recommend the authors to improve the discussion and interpretation of their results in order to better transmits their message to the readers. Hopefully, the major and specific comments reported below will help the authors to improve their work.

MAJOR COMMENTS

I have the strong feeling that the manuscript is overloaded with figures, especially multi-panels figures. First of all, due to the similarity between PM1 and PM10 (Figure 1, 2, 4), the discussion and presentation of results become particularly redundant in Section 3.2, 3.3, 3.4. The subsequent effect is that the discussion often focusses on the differences between the two aerosol fractions rather than on the reasons leading to the multi-year trends or seasonality. I thus suggest the authors show and describe PM10. This will lighten the paper and give more space for the climatologic interpretation of the results. Moreover, it appears that a considerable number of figures is poorly described or is not essential to the understanding of the results. I thus suggest the authors to reconsider the absolute relevance of certain graphs and to remove them from the manuscript or move them to the supplementary. More details can be found in the specific comments.

The dataset allows the investigation of multi-year variability and trends of AOPs and size distribution. The variability of AOPs is also investigated on a shorter time resolution but ignoring the year-to-year variability (Section 3.4, 3.5, 3.6). Thanks to the long-term measurement I would expect a work focusing on trends and multiple-year variability of AOPs. However, the analysis of trends is disconnected from the seasonal and diurnal variability and the consequent RFE. Therefore, I have some troubles in understanding what is the topic or scientific question acting as a glue between the sections, which in some cases (Section 3.3.1, 3.5 and 3.6) appear to be self-standing. I would thus suggest the authors to better exploit their long time series and focus on the long-term evolution/variability of AOPs including trends, impacts on seasons and, potentially, diurnal variability. For instance, Section 3.3.1 is based on 2 months measurements only, what is the long-term implications of NPF on the aerosol optical properties, and is this short period representative of the 10 considered years? Moreover, Section 3.5 provides the diurnal variability of AOPs. Despite the fact of a weak variability, was the boundary layer dynamic changing within the 10-year period? It is hard to understand the relevance and implications of such variability. Similar reasoning applies to the monthly variability, did summer and winter experienced a change from 2006 to now?

The calculation of the forcing efficiency is an extremely interesting topic, and up to me a decadal trend of RFE might represent the core of the entire manuscript together with Section 3.2 and 3.3. However, for the RFE estimations, the authors assumed the atmospheric (RH, cloud) and environmental (surface albedo, day length) variables as constant, which are not even specific for the SMEAR II station. Due to strong seasonality and, potentially, year-to-year variability of such variables, the final RFE estimations are unrealistic. I would suggest the authors implement constants representative, at least, of Southern Finland or, better, to use seasonal dependent variables. Other than that, any conclusion on climatic impacts of aerosol at SMEAR II will be highly questionable and of low interest.

SPECIFIC COMMENTS

P3L11: why MAAP and PSASP are introduced if only data from the AE31 are used?

P3L13: this is irrelevant for the present manuscript.

P3L23: I would say that, since Luoma et al. 20xx is not available, a better description should be provided here.

P3L31: can the author exclude the influence of hygroscopic growth?

P4L4: which instruments, all of them?

P4L11: is not clear why the truncation correction was not applied for the back-scattering. Does it mean that back-scattering can be affected by systematic error compared to total scattering? Was this assessed? Was it negligible? The authors present more than 10 years of data, more care in the presentation of the data correction is mandatory.

P4L19: multiple correction procedures were used or only the Collaud Coen et al. (2010) as stated later? If the correction of Collaud Coen was used, I honestly do not see the reason to cite all the other algorithms. Generally, I would not recommend the frequent self-citation of works that are not ready yet.

P6L15: I expect that BC from biomass burning and traffic has a different chemical composition. Isn't it in contrast with lines 13-15? If AlphaAbs is simultaneously affected by size, chemical composition, sources and mixing, to what purpose is AlphaAbs used here?

P6L22: Equation 5 is quite different from Haywood and Shine (1995), is this the original source of the equation? What is the wavelength of RFE? Is then the aerosol optical depth measured or everything is calculated from Equation 5? Though I have quite some doubts on the choices of constants (see comments on Section 3.7), a better description of the equations and its limits should be provided, together with the motivations at the base of the choices of the constants and the subsequent uncertainties.

P10L5-17: this part of the section mostly describes the technical aspects of the measurements. I would suggest to move them in the method section. Potentially into a new subsection discussing the data coverage and how the data set was reduced/validated.

P11L3: this is the first and last time $\omega 0$ was discussed in Section 3.3. I am wondering if the four panels in Figure 4 showing $\omega 0$ are needed at all.

P11L21-24: the inverse proportionality between ïĄąsca and GMD is supposedly caused by the bimodal size distribution of the aerosol and the substantial presence of accumulation particles. Despite supported by a reference, there is no direct explanation of the physical causes behind such proportionality. Since this is contrary to expectations, as stated by the authors, a deeper reasoning and explanation should be provided.

P12L7-10: The diameter of the particles is the driver for both AlphaSca and b, I have some difficulties in understanding the relevance of the findings described here.

P13L2-3: here is stated that long-range transport brings pollution to the station, but 70% of black carbon comes from local and regional sources (P13L7). These two statements are contradictory.

P13L4-5: I am not sure to understand the relevance of the polar dome here.

P13L30-33: you have the size distribution data, why should you make a hypothesis on size distribution from optical properties?

P15L15-18: Here you need to be careful with the instrumental error. Do you mean that absorption was close to the detection limit of the instrument or that dominant presence of non-absorbing particles caused a decrease of light transmitted through the filter and apparent absorption (Müller et al., 2011)?

P15L23-24: the RFE trends are not described, discussed or interpreted. This recalls my major comments. The manuscript is loaded with data that are never discussed. Provide an interpretation or remove Fig.10a. By the way, add to all figures the panel reference.

P15L26-28: since RFE is calculated from b and w0 and all the environmental variables are kept constant, RFE must change with b and w0. As follow up to the second major comment, the authors are required to provide a deeper interpretation of their results.

P15L29: how the monthly RFE should be interpreted if the atmospheric and environmental parameters are kept constant? Moreover, it appears that the constants are not representative of SMEAR II. So, what should we really learn out of RFE?

P16L5-8: the problem here is that the aerosol optical depth is affected by RH and subsequent hygroscopic growth. So, all your RFE are systematically underestimated by an unknow factor. However, it is unclear if optical depth is measured or calculated.

P16L9-10: Nessler et al. (2005) suggested that water uptake does not enhance absorption coefficient of BC.

P16L11-13: from this work, it is impossible to quantify the change of radiative forcing, nor the effects on the climate. First, RFE trends are not discussed: Second, the absolute values of RFE, as admitted by the authors, are far from being realistic. Moreover, why should we use RFE as “an indicator of how the properties of the aerosol particles have been changing” if the changes of aerosol particles have been measured (Section 3.2, 3.3, 3.4)?

P17L1-2: Fig 1 and Fig. 2 show a net decrease of aerosol number concentration, but is this due to the implementation of new emission policies only? How did precipitation and air circulation changes from 2006? I would recommend the authors to consider all possibilities and base their final conclusions on their data and existing literature.

F1: this figure is too crowded, I do not think that showing both PM1 and PM10 as any relevance (see major comments).

F2: here 5 panels are used to show that the total number of particles decreases and the size distribution is shifted to the smaller diameters. I would say that two panels will do efficiently the job. For example, one panel showing the total particle number concentration (Nfine+Ncoarse) and a second panel showing the ratio between Nfine and Ncoarse or the GMD. Note that Nfine is never defined in the text, is this accumulation+Aitken+nucleation? Please provide a description.

F1-2 As a follow up of my previous comments, I would find a way to merge together a reduced version of Figure 1 and 2, with the goal to focus on the relationship between physical and optical properties described in the text.

F3 This Figure is mentioned only once at P9L26, it does not appear to provide a key

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insight into the understanding of data interpretation. I would thus recommend to move it to the supplementary.

F5: the size distribution of PM10 contains all the necessary data to investigate the size distribution in PM1. This is clear in panels (c) and (d), where the size distributions below 1um are exactly the same. This recalls my general comments, is a separated discussion of PM1 and PM10 really necessary?

F6: The figure shows the nucleation events and the related change in the real part of the refractive index. However, I think that it is largely overcrowded. On page 12, lines 26-27 sufficiently describe the absence of change in the observed AOPs. Due to the low relevance of AOPs variability in this context, I would suggest removing the third and fourth panels from the top. Finally, I am wondering what is the relevance of 2 months data over a 10 year period.

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