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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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GENERAL COMMENTS

This manuscript reports on the optical and microphysical properties of aerosols collected for over a decade at the SMEAR-II atmospheric monitoring station in Finland. These data are valuable in determining long-term trends and variabilities in aerosol properties, which are useful to climate modelers. The statistical distributions of aerosol properties presented in this paper should also be useful to GCM and CTM modelers for model initialization and validation exercises. As such, I think this paper is appropriate for inclusion in ACP and warrants publication after attention to the comments listed below.

The paper is well organized and there are only a few places were the English usage could be improved. The methodology used by the authors is excellent and of high

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quality, and the data presented are in general valid and relevant. I do have some comments that suggest some additional thought be given to provide better explanations of the observations, and I think a better discussion of how drying the sampled aerosols might influence the RFE results is warranted (see comments below). The paper is a bit long but the amount of data being presented from over a decade at this site and the necessary discussions warrant a longer paper. In looking for possible ways to decrease the length of the paper, the only thing I see is to remove the size distribution discussion. While it is interesting in its own right and assists in the interpretation of the AOP data, it is not strictly necessary in this paper. I will leave that decision up to the authors and the editor.

In order to better interpret trends and variability, some estimates of the measurement uncertainty should be provided. I would point the authors to the work of Sherman et al. (2015, ACP), who put a great deal of effort into estimating measurement uncertainties for aerosol optical properties. There is no need to repeat this exercise in detail, but at the very least this reference should be included and some mention of the measurement uncertainties for the TSI nephelometer should be provided.

It was a bit disappointing to find little or no discussion in sections 3.1-3.3 on the relevance and importance of the measurements and their long-term trends and variability. A considerable amount of discussion is presented in sections 3.4-3.7 to explain the seasonal and diurnal variability, etc., and I would like to see more of this in sections 3.2 and 3.3. For the trends, for example, it would be useful to know how these trends compared with other long-term trends in Europe. The Pandolfi paper is cited and is an excellent place to start. Some additional information can be found in Collaud-Coen et al. (2013, ACP), and this paper should also be cited when comparing the optical property (scattering and absorption) measurements. There is also little discussion on the importance of measuring the optical properties in two different size ranges (PM1 and PM10). What does it tell you about sources, ageing, human contribution, etc., if the PM1 fraction for a given parameter is almost as large as the PM10 fraction? What

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does it mean if the PM1/PM10 ratio is changing over time? The authors went to the trouble of adding this additional set of PM1 measurements in 2010 and have several tables and graphs in this paper showing the results. They need to say why they are important and what we learn from them.

The purpose of the lengthy discussion of the comparison of the optical and microphysical properties on page 11 is not clear to me. The manuscript title indicates that this manuscript is about the aerosol optical properties, so why are there size distribution data included in the results and discussions? They are of course useful for interpreting the optical properties, so they have value, and good agreement between measured optical properties and ones calculated from the microphysical measurements give increased confidence in the findings of the study. Perhaps the authors can state that more clearly. The size distribution results could also go in the supplemental materials section if length of the paper becomes a concern.

The RFE calculations in this paper use the global average constants of Haywood and Shine rather than ones estimated or derived for the local area. This is probably OK for trend analysis but the magnitude of the forcing is wrong, especially when considering seasonal variations. For example, the constant used for the global average surface albedo (0.15) does not represent that of the boreal forest around SMEAR-II station over all seasons. . . it should be significantly higher in winter due to snow cover and (I would guess) lower in summer. Also, the measurement relative humidity (RH) and the ambient RH were generally different (this occurred most frequently in the summers). The authors state that if the sample RH was above 40%, the data were flagged and marked as invalid. This implies that the SSA and b values are only accurate when the ambient RH was low (i.e., close to the measurement RH), and that the RFE results are only appropriate for times when the ambient RH was low. Aerosol hygroscopic growth is generally thought to increase the ambient light scattering coefficient much more so than the ambient light absorption coefficient, which would lead to a higher single-scattering albedo and, most likely, a more negative top-of-the-atmosphere RFE value

(i.e., stronger cooling effect). If the ambient RH was higher in many cases than the measurement RH and these measurements were removed from the data set, the reported data set is biased toward a smaller (less negative forcing) cooling effect. Given that the RFE values are most likely not representative of the SMEAR-II region (they use the global average constants) or actual atmospheric conditions, I question their value in this manuscript. If they are to be kept, the authors should re-emphasize that the RFE results are technically meaningful only in the trend analysis (in Table 3) and that the calculations are for dried aerosols using global average constants and thus considerable caution should be used when trying to interpret seasonal variation in RFE at SMEAR-II (Fig. 10). The RFE results could also be moved into the supplemental materials if length of manuscript is a concern.

SPECIFIC COMMENTS

Pg. 1, Line 14: Replace the words 'affected to' with 'influenced'.

Pg.1, Lines 20-21: 'For the aerosol particles to have a cooling (warming) effect, the reflectivity of the particles must be higher (lower) than the albedo of the surface. . .'. What is the definition of 'reflectivity' the authors are using (or is it being used in a qualitative sense here)? For aerosol particles, are the authors referring to aerosol single-scattering albedo (SSA) or some other reflective properties of the particles? It is not technically correct to state that '. . .the aerosol particles . . . have a cooling (warming) effect (if) the SSA of the particles (is) higher (lower) than the albedo of the surface. . .'. Solar photons can be elastically scattered in the forward direction, which does not appreciably cool the surface or lower atmosphere. I would recommend removing this sentence as it is not really necessary anyway, but if kept in the manuscript the authors should state how they are defining the term 'reflectivity' and how that is being compared to surface albedo.

Pg. 2, Line 6: Replace 'concentration' with 'mass and/or volume'. Extensive AOPs are not dependent on the concentration of the particles but on the amount of aerosol

present. Freshly formed particles may have extremely high concentrations in the atmosphere and show very low scattering values.

Pg. 2, Line 8: Replace 'concentration' with 'amount of aerosol'. Same explanation as above.

Pg. 2, Line 11: Eliminate '. . . and not only on the amount of scattering and absorption.'

Pg. 2, Lines 27-28: Why is it important to measure the AOP's of PM1 particles? This should be stated in the manuscript somewhere.

Pg. 3, Line 13 and Line 23: When will the Luoma et al. manuscript in preparation be available? Will it be available by the time this manuscript is published? If not, other references on how the various instruments compare would be appropriate.

Pg. 3, Lines 19-22: The reported AOP's will vary depending on the measurement conditions. The direct aerosol radiative forcing effects at SMEAR-II, however, depend on the ambient conditions of T, P and RH, which were not usually the same as the measurement conditions. A discussion of how this would affect the results is appropriate. Are your seasonal results biased by a) eliminating the high ambient RH periods (which occur more frequently in the summer) before the driers were installed in 2013, or b) accepting these periods after 2013 with high ambient RH but reduced measurement RH? Some discussion of the fraction of data flagged as invalid due to high ambient RH before 2013 is warranted, as is the fraction deemed acceptable (with significant drying) after the driers were installed. This way the reader can understand if this was a frequent or merely occasional occurrence.

Pg. 4, Line 2: How warm does the sample air to the APS instrument get? Does this heating to above room temperature remove any volatile species other than water (e.g., ammonium nitrate)?

Pg. 4, Lines 11-12: 'We did not apply the truncation correction to the backscattering, since the backscattering measurements were much noisier, especially at the red

wavelength.' OK then the determination of b is wrong, as is the calculation of the up-scatter fraction, and the question is how far off are your values from the fully truncation-corrected values. An estimate of the uncertainty or error that enters the calculation of b due to not applying the truncation correction to the sigma-bsca values should be given. I agree that the sigma-bsca values are quite noisy at 1-minute resolution. At what resolution were you recording the raw data (1 second?,1 minute?, 10 minutes?, I don't see this listed in the manuscript)? Could you have averaged the sigma-bsca values to hourly or longer resolution before applying the corrections? This would perhaps help to beat down the noise a little.

Pg. 4, Lines 18-23: Which algorithm(s) or recommendations in Collaud Coen et al. (2010) were used? In that paper they evaluated four previous aethalometer correction schemes (Weingartner, Arnott, Schmid and Virkkula) and they also made new recommendations on the applicability of each in different circumstances.

Pg. 5, Line 12: Replace the word 'direction' with 'hemisphere'.

Pg. 5, all equations: The subscript font is quite small. Possibly it will look better in the published version.

Pg. 4-6, Section on Data Processing: Somewhere in this manuscript the authors need to give some estimate of the measurement uncertainties of the instruments they are using. I recommend looking at the work of Sherman et al. (2015, ACP) to see how they calculated the measurement uncertainties. It is a lot of work so I do not recommend that you try to repeat those analyses, but you should be able to reference their Table S2 'Total and precision fractional uncertainties (%) of measured PM1 and PM10 aerosol optical properties (AOPs) σsp, σbsp, and σap and calculated AOPs (e.g., the intensive AOPs) for 1-hour averaging time. Uncertainties are expressed as 95% confidence intervals.' and state the uncertainties relevant to your report.

Pg. 6, Line 8: '. . .the absorption would be dependent on wavelength as lambdaˆ-1. . .'. Rephrase as '. . .the absorption would have a wavelength dependence of approximately

lambdaˆ-1. . .’.

Pg. 6, Lines 28-29: While adjusting the AOP’s to a common set of conditions is appropriate (and indeed necessary) to evaluate trends and to compare properties at different sites, you need the measurements at ambient conditions to determine the effects of aerosols on perturbing the surface radiation balance (i.e., their direct radiative/climate forcing effect). It would be good to provide some estimate or limit as to how different the AOP’s are for dried vs. ambient air. Perhaps an example calculation, where the AOP’s are adjusted to ambient conditions using some assumed conditions of T, P and RH, would help. I am sure there are studies of Finnish/Scandinavian/northern European aerosols where the aerosol hygroscopic growth was measured or calculated. These results could be used as a very rough scaling factor to calculate the AOP’s at SMEAR-II at ambient atmospheric conditions. Otherwise the reader will not know if the presented dry aerosol RFE results are even close to those for real atmospheric conditions at SMEAR-II.

Pg. 7, Line 15: Replace ‘describes’ with ‘provides information on’.

Pg. 8, Line 3: Replace ‘chapter’ with ‘section’.

Pg. 8, Line 15-16: ‘Naturally, the different methods used in the absorption data processing also affected the optical properties, which are dependent on the sigma-abs, such as omega-0 and k.’ How much of a difference in omega-0 or k can be attributed to the different data processing methods? Is it a large or small difference? Could you provide an example where the same processing is used in two different time periods that shows how large of an effect this is?

Pg. 9, Line 6: Replace ‘marked’ with ‘included’.

Pg. 9, Section 3.2, second paragraph: The 13%/year decrease in the ïĄşabs value at SMEAR-II is an important finding and should be emphasized here! Has this been observed at other sites in Finland and/or Europe? Can you provide a hypothesis as

to why this happened over the last decade at SMEAR-II station? Could it be more local or regional/continental scale effects? Is it due to less soot aerosols? Or possibly decreasing amounts of BrC?

Pg. 13, Line 3: Replace 'means' with 'suggests'.

Pg. 14, Section 3.6, second paragraph: The difference in the PM1/PM10 scattering ratio between Virkkula (2011) at 85% and the current study at 75% is a little concerning. There could have been long term changes in the environment at SMEAR-II region that might partially explain this, or it could be a difference in sampling conditions. Was there any RH measurement made at or near the impactors (as opposed to inside the nephelometer)? You need an RH measurement taken near the impactors to ensure you have a proper size cut (i.e., without the possible artifact you mention).

Pg. 15, Section 3.7: It needs to be stressed that the RFE calculations are for dry or semi-dry (RH<40%) aerosols.

Pg. 15, Line 23: '. . ., which makes the RFE decrease.' After decrease, add the parenthetical phrase '(i.e., become more negative)'.

Pg. 15, Line 26: Replace 'ine' with 'in'.

Pg. 16, Lines 5-13: This is a good explanation! The authors state that while the magnitude of the RFE perturbation cannot be precisely determined using this methodology, the trends probably can, and the RFE estimates they provide are most likely a lower limit to the true cooling effect.

Pg. 16., Lines 18-23, and Fig. 12: This is a discussion of systematic variability of aerosol optical properties. This type of systematic variability has been observed before. The earliest paper I know of that discussed this was Delene and Ogren (2002, J. Atmos. Sci, Fig. 8) which should be referenced. This was also in Sherman et al. (2015, ACP, Figs. 10a, 10b, 10d). Their results are consistent with those presented in this paper.

Pg. 27, Fig. 3: The largest decrease over time is for the larger accumulation mode

particles (i.e., 0.4-0.7 micrometer diam). Any ideas why?

Pg. 29, Fig. 5: The text in the legends are very small. This may, however, be acceptable to the technical editor.

Pg. 31, Fig. 7: Why are there breaks in the whiskers and some whiskers not attached to the boxes? Is this a plotting artifact or is additional explanation necessary as to what the whiskers are meant to display?

Pg. 33, Fig. 9: It appears that the whiskers are drawn as dashed lines with relatively long dashes and breaks. These should either be changed to solid lines or else changed to broken lines with smaller breaks in them.

Pg. 35, Fig. 11: Caption '. . .1000 grid points in total.' Should this be '10,000 grid points in total.'?

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