RESPONSE BY THE AUTHOR

First of all, than you for your comments! They were of great help in improving this study. I first reply on some major comments you both had and then, I reply to each of the comments separately.

1 MAJOR CHANGES AND RESPONSE TO COMMON COMMENTS

1.1 SHORTENING THE MANUSCRIPT

We analyzed a long data set in this study and we presented a lot of figures in the first manuscript. To emphasize the important parts of the manuscript, such as the trend analysis and RFE, we removed some of the figures as you recommended. For example, we got rid of the PM1 panels on some of the figures, since they gave no extra information that was relevant for this study. We also removed the section about the new particle formation events and aerosol optical properties. We agreed that it did not fit in the topic of this manuscript. I moved the old Fig. 3 to the supplementary material, since it was referred only once in the text. I moved the old Sect. 3.5 (Diurnal variation) to the supplementary material. The diurnal variation was a bit separate from the rest of the manuscript and it did not present any new information to Virkkula et al. (2011). I also moved the old Fig. 11 to the supplementary since after adding the seasonality and ambient RH to the RFE analysis, this figure did not feel important anymore. The number of figures decreased from 12 to 8 and the number of subfigures decreased as well. However, the supplementary material grew from one figure to nine figures since we answered to some of your questions without increasing the number of figures in the main article.

Since we added more description in the method section and improved the discussion we got now more text. We also added one table, which describes how the trend varies between different seasons.

1.2 SIZE DISTRIBUTION ANALYSIS

We are still keeping the size distribution analysis, since we found it interesting to study how the aerosol optical properties and their trends are related to the size distribution. For example, in Pandolfi et al., (2018) they were not using size distribution data and they had to assume what kind of changes in the size distributions cause the different types of trends for the α_{sca} observed at different stations. The study on the size distribution helps us to understand how the size dependent b and α_{sca} vary between PM1 and PM10 measurements. However, we moved part of the size distribution analysis (old Fig. 3) to the supplementary material.

1.3 CALCULATION OF RFE

Another thing you both pointed out was the calculation of RFE by using global average values. We worked more with the topic and determined a more realistic RFE at SMEAR II. What I found difficult was determining the b for moist conditions and I had to make rather rough estimates there. This would be an interesting topic to study more.

To emphasize the meaning of RFE: as stated by Sherman et al. (2015), the RFE ($\Delta F\ \delta^{-1}$) provides a means for comparing the intrinsic forcing efficiency of aerosols measured at different sites. The RFE describes the change that aerosol particles would have on the top-of-atmosphere radiative forcing (ΔF) per unit of aerosol optical

depth (δ). Since AOD is unitless, the unit of RFE is W/m2. The RFE is an intensive property and it does not depend on the amount of aerosols. If we wanted to know the Δ*F*, we would need measurements of δ, which, on the contrary, is an extensive property and depended on the amount of aerosols.

We have now determined three different types of RFE values:

1) $RFE_{H\&S}$ was calculated by using the constant values suggested by Haywood and Shine (1995). $RFE_{H\&S}$ was derived for dry particles. Here the subscript "H&S" refers to Haywood and Shine (1995).

2) RFE_S was calculated by using seasonal averages for the environmental parameters (D, A_C, R_S). So here we let the fractional day length to vary; we used more realistic values for the surface reflectance according to Kuusinen et al., (2012) and took the snow cover into account; and we determined an average cloud fraction for each month. More detailed description is provided in the manuscript in Sect. 2.3.3 (p. 8 – 10). We added a figure in the supplementary material (Fig. S1a-c) describing the seasonal variability of these parameters. RFE_S was derived for dry particles. Here the subscript "S" refers to "seasonal".

3) $RFE_{S,moist}$ was calculated similarly to RFE_S, but taking the ambient RH into account. Here we determined average RH for each month and we derived the AOPs to the average humidity. The seasonality of RH is presented in Fig. S1d. In determining the ω_0 for humid conditions, we assumed that the absorption does not depend on the RH. The scattering was converted to humid conditions using the parametrization provided by Zieger et al. (2015), which is presented in Sect. 2.3.1. The parametrization was given only for total scattering so we could not use it for backscattering and determine b with this parametrization. Instead we assumed that the b has a linear dependency on RH. Fierz-Schmidhauser et al. (2010) observed that the b decreased 30 % when the RH increased from dry conditions to 85 %, which we used in this study. Here the subscript "S,moist" refers to "seasonal" and ambient RH, which was > 50 % for each month. This is described in Sect. 2.3.3 (p. 10).

The results for different RFE values are presented in Sect. 3.6, Table 1., and Fig. 7.

1.4 DATA PROCESSING

You both commented that I should apply the truncation correction also to backscattering data. This has now been done.

I also made a small change to the Aethalometer flow correction (Fig. S2 added in the supplement), and changed the C_{ref} value from 3.35 to 3.19. Thus there are small changes (less than 5 %) in the data presented in the article.

In this new version of the manuscript we have discussion about how the RH affects the scattering. Therefore we present the parametrization of the scattering enhancement factor in the Sect. 2.3.1. I have also added a better description about the Aethalometer correction algorithm used here. Since we were not able to submit my second manuscript, which would have presented the C_{ref} at SMERA II, I added a short description about C_{ref}.

2 RESPONSE TO MAJOR AND SPECIFIC COMMENTS

The comments by the referees are listed with bolded font and the response by the authors are written with normal font. If the answer refers to a text that was added in the manuscript, the quotation is italicized.

2.1 REFEREE 1

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 background 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 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.

We have now worked with the manuscript and improved the discussion especially in the sections that concern trend and RFE. We have also decreased the number figures in order to make the manuscript more readable and less crowded.

MAJOR COMMENTS

I have the strong feeling that the manuscript is overloaded with figures, especially multipanels 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.

I have now modified the multipanel figures so that they include only PM10 data, if the PM1 data presented no additional information. Some of the figures, which did not seem that relevant anymore, I have moved to the supplementary material.

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?

There is now a figure in the supplementary material (Fig. S3) that better describes the year-to-year variability of scattering, absorption and particulate volume for each season. The figure presents the time series of monthly medians separately for each season. It can be seen from the figure that the year-to-year variability is the highest in winter, when the amount of pollution is highly depended on the meteorological conditions, which is discussed in the supplementary.

To connect the trend analysis to the seasonal variation we determined trends separately for each season. The results are presented in Table 4 of the manuscript.

I agree with the Sections 3.3.1 and 3.5 being self-standing and we chose to remove 3.3.1 from this manuscript. Sect. 3.5 I chose to move in the supplementary.

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.

We have now worked more with this topic. See my answer 1.3 in the beginning of this document.

SPESIFIC COMMENTS

P3L11: Why MAAP and PSAP are introduced if only data from the AE31 are used?

I removed PSAP from the introduction but I left MAAP since I added a short description about the multiple scattering correction factor (C_{ref}) (see my response to comment about P3L23), which was determined by comparing Aethalometer and MAAP measurements.

P3L13: This is irrelevant for the present manuscript.

I removed this sentence.

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

I removed the self-citation from the manuscript. We had too optimistic expectations about the timetable with the other manuscript. We hope to submit the manuscript by the end of this year. I have also added a better description about determining the C_{ref}:

"The C_{ref} was determined by comparing the Aethalometer data, that was corrected only for the filter loading artefact, against the reference absorption coefficient ($\sigma_{abs,ref}$) measured by the MAAP.

$$C_{ref} = \frac{\sigma_{ATN}}{L \cdot \sigma_{abs,ref}}. \quad (6)$$

The resulted median value for C_{ref} was 3.19, with a standard deviation of 0.67. "

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

When the particles enter the electric charger, the sample air is not dried. However, since the sheath air is dried, the aerosol particles are also dry in the DMA of the DMPS setup. I added this explanation to the manuscript:

"There is no active drying system in the TDMPS sample line to prevent particle losses. However, the sheath flows, which are used in the TDMPS system, are dried (RH < 40 %) so the particles are sampled in dry conditions."

P4L4: Which instruments, all of them?

I meant only instruments that measure optical properties. For other instruments we should not have this problem. I fixed the sentence to:

"In the present work, if the internal RH in any of the optical instruments exceeded 40 %, the data from that instrument were excluded from further analysis if not stated otherwise."

P4L11: It is not clear why the truncation correction was not applied for the backscattering. 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.

Now, also the backscattering is corrected.

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.

I removed the self-citation and the citations to the other correction algorithms that were not used here. I added a description of the correction algorithm in the text:

"Here, we corrected the Aethalometer data by using the correction algorithm described by Collaud Coen et al. (2010)

$$\sigma_{abs,i} = \frac{\sigma_{ATN,i} - a_{s,i}\bar{\sigma}_{sca,s,i}}{C_{ref}\, L_{s,i}}, \quad (3)$$

where

$$L_{s,i} = \left(\frac{1}{l(1-\bar{\omega}_{0,s,i})+1} - 1\right) \cdot \frac{ATN_i}{50\ \%} + 1, \quad (4)$$

and

$$a_{s,i} = \bar{\zeta}_{sca,s,i}^{d-1} \cdot c \cdot \lambda^{-\bar{\alpha}_{sca,s,i}\cdot(d-1)}. \quad (5)$$

In Eqs. 3 and 4, the subscript i indicates the number of the measurement and the subscript s indicates the average properties of the aerosol particles that are embedded in the filter spot. The over lined parameters are mean values from the start of the filter spot to the ith measurement. In Eq. 3, the σ_{ATN} is the attenuation coefficient reported by the Aethalometer, a is the scattering correction parameter, C_{ref} is the multiple scattering correction factor, and L is the loading correction function. In Eq. 4, the ω_o is the single scattering albedo (see Sect. 2.3.3) and the ATN is the light attenuation through the filter spot in percentages. In Eq. 5 the ζ_{sca} is the proportionality constant of the wavelength power law dependence of σ_{sca} and α_{sca} is the Ångström exponent of the σ_{sca} (see Sect. 2.3.3). For l, d, and c we used values 0.74, 0.564 and 0.329 $\cdot 10^{-3}$ respectively. For scattering correction, we used measured σ_{sca} values that were interpolated and extrapolated to the AE-31 wavelengths. Note that most of the symbols used for the variables are different from Collaud Coen et al. (2010). The reason is that in the present work the symbols are used for other variables below."

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 α_{abs} is simultaneously affected by size, chemical composition, sources and mixing, to what purpose is α_{abs} used here?

I guess the Aethalometer model assumes that the α_{abs} is only depended on the chemical composition, which then depends on the source. In Hyytiälä, the BC particles are typically aged so they probably have a coating that affects the α_{abs} and because of this the Aethalometer model might not be functioning in Hyytiälä. We added some discussion about the aethalometer model in the discussion section:

"Also, the α_{abs} is typically associated with the source of the BC and it is often used to quantify whether the BC is traffic or wood burning related (Sandradewi et al., 2008; Zotter et al., 2017) so that high α_{abs} is a sign of wood burning. In the source apportionment, α_{abs} close to one indicates that the BC is sourced from traffic. Since we observed relatively higher α_{abs} in winter, the results are in line with the assumption of domestic wood burning that takes place during winter. However, in summer, α_{abs} was often < 1, which would yield an unphysical fraction (over a 100 %) of traffic related BC. Values below 1 could have been caused by large BC particles (D_p > 100 nm) that have a purely scattering coating (Lack and Cappa, 2010). It must be noted that the α_{abs} depends also on the correction algorithm. For example, if the σ_{abs} was corrected with the algorithm proposed by Arnott et al. (2005), the mean ± SD of α_{abs} would have been 1.36 ± 0.51 (see Table S2). Using the α_{abs}, which was determined by using the correction by Arnott et al. (2005), the results for the source apportionment would be different and they would show higher fraction of BC from wood burning. Further investigation of the complex nature of α_{abs} is omitted 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.

It is the same equation as the Eq. 3 in Haywood & Shine (1995) but divided by the AOP. This was derived by Sheridan and Ogren (1999) (Eq. 8), to which I was missing a citation. Here we have not used the measurements of AOD. Haywood & Shine used the constants for calculating the ΔF independent of wavelength and Sheridan and Ogren (1999) used these same values in calculating the RFE at 550 nm. In addition to the H&S constants, we now calculated the RFE by using seasonally varying environmental parameters (see my 1.3 at the beginning of this document). Using seasonally varying parameters give some estimate about how realistic the RFE calculated by using the constant values is.

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.

I moved this part to a new section "2.3.4 Data coverage" and described the data coverage better. I also added a table in the supplementary material where the data coverage from each month is presented.

"If averaged over the whole measurement period, 81 % of the nephelometer data and 70 % of the aethalometer data were considered valid. All the AOPs had some gaps in the data (see Fig. 1). Most of these gaps in the time series of AOPs during the summers of 2009 and 2010 were due to too high RH. The gap in 2010 was due to maintenance and installation of the dryers and the switching inlet system. Some additional σ_{bsca} data were missing, due to malfunction of the backscatter shutter of the integrating nephelometer. Dirty optics, malfunctions and maintenance caused the gaps in the σ_{abs} data in 2012 and 2015.

Until March 2010, the integrating nephelometer and the aethalometer measured sample air that was not dried with any external dryers. During winter, the relative humidity (RH) remained below 40 %, since the sample air warmed up to room temperature (about 22 °C). Sometimes in summer, the RH of the sample increased to over the 40 % limit. If the RH was above 40 %, the data were flagged as invalid and they were omitted from the data analysis if not stated otherwise. About 25 % of all the data before March 2010 had to be removed due to too high RH. Almost all of the removed data was from summer and fall months (June – October) and if regarding only these months, 46 % of the data were flagged. If the moist data was included the overall data coverage would increase to 89 % and 77 % for scattering and absorption data, respectively. Monthly data coverage is presented in Table S1. After the installation of the Nafion-dryers in March 2010, the humidity caused no further problems."

P11L3: This is the first and last time ω_0 was discussed in Section 3.3. I am wondering if the four panels in Figure 4 showing ω_0 are needed at all.

We added discussion concerning the ω_o panel:

"Kulmala et al. (2016) estimated that fresh eBC particles observed at SMEAR II are in the size range of 80 – 120 nm. That estimate was calculated in a simplified way from the relationship between particle number

concentrations and BCe concentrations. A better estimate is obtained from the size dependence of ω_o. The darkest aerosol has ω_o < 0.6 and GMD in the range of about 30 – 70 nm (Fig. 3b, 3f, and 3j). This has been shown to be the range of fresh BC (e.g., Kittelson, 1998; Casati et al., 2007; Zhang et al., 2008) which suggests the source of BC is not far, probably within some kilometers only."

P11L21-24: The inverse proportionality between α_{sca} 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.

This was more and a detailed explanation is given in the Sect. S6 in the supplementary material. This was also described in the main text:

"To study the reasons behind this relationship we generated first unimodal size distributions with two geometric standard deviations GSD = 1.5 and 2.0 and calculated both σ_{sca} and σ_{bsca} at λ = 450, 550, and 700 nm with the Mie code with m = 1.517 + 0.19i and the α_{sca} and b from them. For unimodal size distributions the α_{sca} decrease with increasing GMD as is shown by the lines in Fig. 3c. Schuster et al. (2006) showed that the relationship may be the opposite for bimodal size distributions. Schuster et al. (2006) explained this behavior by that adding a larger or coarse particle size mode to a fine particle mode that is inefficiently scattering - for instance nucleation and Aitken mode particles – the larger mode contributes more efficiently to the Ångström exponent than the fine mode. The contribution of the particles smaller than 100 nm to GMD is larger than that of the larger particle modes which leads to the observed relationship. To study this in more detail we generated also bimodal size distributions. The analysis presented in the supplement (S4) shows that the α_{sca} of bimodal size distributions can be calculated as a linear combination of the α_{sca} of the modes, weighted by the fractions of σ_{sca} of the respective modes. This explains the increase of α_{sca} with growing GMD."

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

The point here was to show that the *b* and α_{sca} are sensitive to different size ranges and that the bimodal size distribution for PM10 particles can make the examination of *b* and α_{sca} a bit complicated. For PM1 particles the variation of *b* and α_{sca} is easier to understand since the size distribution is closer to a unimodal size distribution:

"There was a negative correlation between the GMD and PM10 b (Fig. 3d) as expected, but the correlation was rather weak. On the contrary, the correlation between the VMD_{tot} and PM10 b was slightly positive (Fig. 3h). The negative correlation of α_{sca} with VMD_{tot} and the positive correlation of b with VMD_{tot} for the PM10 particles indicates that the α_{sca} and b were sensitive to different size ranges. The α_{sca} decreased when there are more coarse particles present, but for the b the coarse particles seem to have no expected effect and the b increased with increasing VMD_{tot}. Fig. 4a. shows that when the VMD > 1500 nm, the peak of $DV/dlogD_p$ in the accumulation mode was much lower and tilted towards the smaller diameters than compared to the situations where the VMD < 1000 nm. This is in line with Collaud Coen et al. (2007), who stated that in the Jungfraujoch data, b was sensitive to particles smaller than 400 nm and that the sensitivity of the α_{sca} was at its maxima for particle diameters between 500 and 800 nm.

For the PM1 particles, the measured α_{sca} and b were well in line with the modeled values (Figs. 3k and l), since the coarse mode particles were removed prior to the measurements, the shape of the size distribution was closer to a unimodal size distribution, and the VMD_{fine} described better how the accumulation mode shifted."

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.

Here, I meant to say that the local and regional BC emissions are also important in winter. I formulated this paragraph to:

"Hyvärinen et al. (2011) observed increased equivalent black carbon (eBC, the BC concentration determined optically from σ_{abs} measurements) concentrations at SMEAR II in winter, when the long-range transport brings pollution from the central and eastern Europe. However, Hienola et al. (2013) estimated that about 70 % of the measured eBC at SMEAR II is emitted from local or regional sources or transported from Finnish cities so also the local and regional emissions have a significant role in the elevated eBC concentrations."

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

I formulated this sentence and removed the reference to polar dome. I meant that the southern air masses are more common at SMEAR II in winter, which brings pollution from central or eastern Europe.

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

I added a figure about the seasonality of the size distribution in the supplementary material so no hypothesis needs to be done.

*"The seasonal variation in α_{sca} and b depends on the seasonal variation in the size distribution of the particles. Both α_{sca} and b were maximal in summer and minimal in winter, suggesting that in summer, the particle population consisted of smaller particles than in winter. Closer investigation on the size distribution, which is presented in Fig. S3 and S4, reveals that in winter, the VMD_{tot} was experiencing it minimum due to a lack of coarse mode particles. This is in contrast with the observation or smaller α_{sca} and b. In fact, the seasonal variation of α_{sca} and b was explained by the seasonal variation of accumulation mod*e and VMD_{fine}, which is a good indicator for the shifting accumulation mode. *In winter, the accumulation mode was shifted towards larger sizes and the median of VMD_{fine} was about 350 nm. In summer the situation was the opposite and VMD_{fine} was about 250 nm."*

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)?

I formulated this part. I meant the apparent absorption caused by scattering and how it causes relative large uncertainty since the scattering is high and absorption is low.

"The deviation of the σ_{abs} PM1/PM10 ratio clearly varied seasonally. In summer, the variation was considerably higher than in winter. In the correction algorithm, which was used for the absorption data (Eq. 3), part of the σ_{sca} is subtracted from σ_{abs} as an apparent absorption (Muller et al., 2011). This subtraction causes relatively high uncertainty when the σ_{abs} is low and σ_{sca} is high like it is in summer. This uncertainty is emphasized for PM10 measurements, since the σ_{sca} is relatively higher than σ_{abs} if compared to PM1 measurements. The uncertainty in the measurements also explains why there were so many values above 1 measured in the PM1/PM10 σ_{abs} ratio."

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

I added more discussion in Sect. 3.6 about the trends and seasonal variation of RFE.

Fig.10a. By the way, add to all figures the panel reference.

Fixed this.

P15L26-28: Since RFE is calculated from b and ω_0 and all the environmental variables are kept constant, RFE must change with b and ω_0. As follow up to the second major comment, the authors are required to provide a deeper interpretation of their results.

We added more discussion about the effect of environmental variables and about the effect of the RH. See my answer 1.3 at the beginning of the document. Taking the seasonality of the environmental parameters into account amplifies the seasonal variation of RFE. The effect of RH is not as pronounced as the effect of using the seasonally varying environmental parameters.

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?

More discussion about this in Sect. 3.6. As stated by Sherman et al. (2015), the RFE provides a means for comparing the intrinsic forcing efficiency of aerosols measured at different sites, this is the reason for calculating it by using the same constants that have been used in other publications.

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 unknown factor. However, it is unclear if optical depth is measured or calculated.

Our study used only in-situ measurements of AOPs and we have not measured the AOD. I have emphasized this in the text:

"The RFE (or $\Delta F\delta^{-1}$) describes only the efficiency of the aerosol particles in cooling or warming the climate per unit of aerosol optical depth (δ). Eq. 11 assumes that the properties of the aerosol particles are uniform in the atmospheric column that is rarely the case in reality. In ambient air, we should also take into account the variability in RH as a function of height. At the top of the boundary layer we typically have RH values close to 100 %. Here, we determined the RFE by using the RH measured near the ground (16 m). The simplified RFE does not give an absolute value for the aerosol forcing, however, it can still indicate how the changes in AOPs affect the climate."

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

I added a citation to this study and took this finding into account when determining the effect of ambient RH on the ω_o and further on to RFE.

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)?

I added here the RFE calculated by using more realistic values (see my answer 1.3). I also improved the discussion. The point of determining the RFE is not to quantify the radiative forcing (for that we would need a lot more parameters, AOD for example). However it describes how the efficiency of aerosol particles to cool (or warm) the climate has changed during the measurement period.

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.

I formulated this:

"The extensive AOPs, as well as the aerosol number and volume concentration, tended to decrease. Our observation was in line with the other studies conducted in Europe and North America that also observed decreasing trends for the extensive AOPs (Collaud Coen et al., 2013; Pandolfi et al., 2018; Sherman et al., 2015), number concentration (Asmi et al., 2013) and aerosol optical depth (Li et al., 2014). This uniform decreasing trend in the amount of aerosol particles suggests that the anthropogenic emissions of particulate matter and gases that take part in secondary aerosol formation has been decreasing in Europe and North America The observed tendency for b and α_{sca} to increase indicated that the particle size distribution was moving towards smaller diameters. A more detailed investigation revealed that the number of larger accumulation mode particles decreased relatively the fastest, which also supports the assumed decrease in pollution."

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

I removed the PM1 column from here.

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.

I removed two of the panels. Now there are panels for total particle number concentration (N_{tot}), total particle volume (V_{tot}) and VMD_{tot}. We kept these parameters since *N* describes well the overall decrease in aerosol particles and *V* describes the amount of optically active aerosol matter. VMD was used instead of GMD, since it is more sensitive to changes in the optically active size ranges.

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.

I see this point, but I could not merge these figures due to technical reasons (the figure became too crowded and the fonts too small). I would also like to keep the optical parameters separated from the size distribution parameters.

F3 This Figure is mentioned only once at P9L26, it does not appear to provide a key insight into the understanding of data interpretation. I would thus recommend to move it to the supplementary.

We removed this figure from the main manuscript according to your recommendation. However, we kept it in the supplementary material since it proves that the relatively highest decrease occurred in the larger side of the accumulation mode and thus supports the observed trends in increasing *b* and α_{sca}.

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 1 um are exactly the same. This recalls my general comments, is a separated discussion of PM1 and PM10 really necessary?

In Figs. 4a and b we used different VMD values as limit values for averaging the size distributions. These values differ for PM1 and PM10 as well as do the *b* and α_{sca}. So if we want to study how the *b* and α_{sca} are related to the VMD, we need to investigate these separately for PM1 and PM10. However, since GMD is practically the same for PM1 and PM10, we now show the average size distribution limited by the GMD for the PM10 only. We also removed the panel where the PM1 AOPs were compared against GMD in the old Fig. 4.

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.

We removed this figure and the section discussing about it from the manuscript, since we realized that it is too much for this article.

REFEREE 2

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 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.

We have now kept the size distribution section since they explain the behavior of AOPs. However, we cut some other elements (Sect. 3.3.1 and 3.5) from the manuscript to shorten it a bit and to emphasize important results.

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.

We cited this work concerning the uncertainty of the Nephelometer. Since they used PSAP in measuring absorption, we determined the uncertainty of the Aethalometer in a similar manner to Backman et al. 2017. We determined that the uncertainty of the σ_{abs} was about 23 %.

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.

We have now added discussion to these sections, especially to Sect. 3.2.

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 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.

We added a motivation to measure PM1 to the introduction:

"The measurements of AOPs were started for aerosol particles smaller than 10 µm in diameter (PM10). The PM10 measurements are sensitive to coarse particles that are typically primary and originated from natural sources, such as soil dust and sea salt. To obtain additional information about submicron particles, parallel measurements of AOPs for PM1 were launched in June 2010. Motivation to measure also PM1 particles is that secondary aerosols (both natural and anthropogenic), and anthropogenic primary aerosols are typically submicron particles. Having measurements for different cut-offs makes the measurements also more comparable between different stations, since stations might use different cut-off sizes."

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.

We have kept the size distribution study in the manuscript since the size distributions explain most of the optical properties. The size distribution measurements are independent from the optical measurements so together they increase confidence to the results. We even added some discussion of the size dependence of single-scattering albedo as wished by the other reviewer. Another addition is an explanation of the increasing Ångström exponent with increasing geometric mean diameter as also wished by the other reviewer. That analysis is in the supplement.

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.

As stated by Sherman et al. (2015), the purpose of determining the RFE is to provide a means for comparing the intrinsic aerosol forcing efficiency of aerosols measured at different sites. We calculated the RFE by using the constant values to have results comparable with other studies in very different types of environments (e.g. Sheridan and Ogren, 1999; Andrews et al., 2011; Sherman et al., 2015; Shen et al., 2018) and to study how the RFE changes with varying ω_0 and b. However, we have now determined RFE also by using more realistic environmental parameters also. See my answer 1.3 at the beginning of this document.

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.

We have now estimated the ω_o and b for ambient RH as well and taken this into account in the RFE calculations. See my answer 1.3 at the beginning of this document for a more detailed description.

SPECIFIC COMMENTS

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

Fixed.

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.

I reformulated this part.

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.

Replaced.

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

Replaced this as well.

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

Eliminated.

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.

We added a motivation to measure PM1 particles in the introduction (see my answer to one of the major comments).

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.

I had too optimistic expectations about the timetable with that manuscript… I have removed self-citation from the manuscript and modified the text.

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.

The RFE does not depend on T or p, since it is an intensive property. But it does depend on the RH and I have now taken this into account in the paper. I added median values of other AOPs including the high RH conditions as well. I also added a more detailed description about the fraction of invalidated data in the new "2.3.4 Data coverage" section.

I now did the trend analysis for a data set, where the high humidity conditions were included. The trends did not change remarkably compared to the data set, where the moist conditions were excluded. I had a typo in the old manuscript concerning the installation of the driers. The driers were installed already in 2010, not in 2013, so there were only four years of measurements without the drier.

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)?

The sample air is heated to 40 °C and for example ammonium nitrate would be evaporated, however, the concentration of ammonium nitrate is very low in Hyytiälä so this should not affect in our study. Alternatively, there are many other volatile compounds at SMEAR II. These compounds take part in the secondary aerosol particle formation and the resulting particles are typically smaller than what we measure with the APS. Thus we believe that heating the sample up to 40 °C has no significant effect here.

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 upscatter 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 σ_{bsca} values should be given. I agree that the s σ_{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 σ_{bsca} values to hourly or longer resolution before applying the corrections? This would perhaps help to beat down the noise a little.

Backscattering data has now been corrected as well.

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.

We used the new recommendation presented in that paper. I added a better description about the correction algorithm we used.

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

Replaced.

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

I have now increased the font a bit for so that it is easier to review.

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) σ_{sp}, σ_{bsp}, and σ_{ap} 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.

We have now added the uncertainty of the Nephelometer in the manuscript and we estimated the uncertainty of the Aethalometer in Sect. 2.3.2. An estimation for the intensive AOPs is presented in the supplement.

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…'.

Rephrased this.

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.

We did more analysis on RFE, where we took the seasonality of the D, R_S, and A_C into account. We also determined the RFE for ambient conditions by calculating the b and ω_0 for moist conditions. See my answer 1.3 for a more detailed description.

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

Replaced.

Pg. 8, Line 3: Replace 'chapter' with 'section'.

Replaced.

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 ω_0 and k.' How much of a difference in ω_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?

Table S2 in the supplementary material presents various absorption depended AOPs that were determined from absorption data that was corrected using the correction algorithm by Arnott et al. (2005). In this correction algorithm we used the same C_{ref} as Virkkula et al. (2011). Compared to Virkkula et al. (2011), we used a corrected spot size and flow, and we invalidated situations when the RH > 40 % (Virkkula et al. (2011) invalidated situation when the RH > 50 %). The RH limits for acceptable data were taken from WMO GAW recommendations. In the GAW guidelines of 2003 the recommendation was to maintain RH < 50% but the limit was later lowered to RH < 40% (WMO/GAW, 2003, 2016).

The absorption at 520 nm was similar for both corrections but at other wavelengths the were differences. At lower wavelengths the σ_{abs} was higher for the data that was corrected by the Arnott et al. (2005) algorithm. At higher wavelengths the situation was the opposite. Thus there was a notable change is in the α_{abs} that describes the wavelength dependency of σ_{abs} and the α_{abs} was 1.4 for the σ_{abs} data that was corrected by the Arnott et al. (2005) algorithm. For other σ_{abs} depended parameters there were no significant differences at green wavelength. We did not do the analysis for the *k* since running the iteration for the whole data set takes a long time and we expected to see no large differences.

Pg. 9, Line 6: Replace 'marked' with 'included'.

Replaced.

Pg. 9, Section 3.2, second paragraph: The 13%/year decrease in the σ_{abs} 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?

I do not think that there has been this steep decrease in other rural or remote stations or at least I could not find any citations. The trend of absorption in PM1 aerosol was calculated for only 7.5 years of data and it is more sensitive to extreme values in the time series. Here I believe the steep slope was caused by few extreme high values measured at the beginning of 2012. I find the trends calculated for the PM10 data more reliable since it is calculated for over a ten year period. I added some discussion about this difference in the results.

"For the PM1 σ_{abs}, we observed a very steep decrease (-12 %yr^{-1}), which was probably caused by very high σ_{abs} measured in January and February in 2012. Also the data gaps in winter 2013 and 2015 could have affected the

trends. The time series, of which the trends were determined for the PM1 measurements, were only 7.5 years long. Trends, which are determined for shorter time series are more sensitive to year-to-year variability. This kind of extreme values can induce relatively large trends, which is why trend analysis for short time series (less than ten years) should be treated with caution."

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

Replaced.

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).

Unfortunately we do not have any RH measurements near the impactor or the inlet. The impactor is inside the measurement cottage so the air has some time to warm up to room temperature (about 22°C). Of course it does not help during the summer, when the temperature outside is similar to the room temperature. The difference that we observe between these two studies could have been caused by some technical issues like this.

Another thing is that Virkkula et al. (2011) determined the ratio from scattering calculated with a Mie model from size distributions measured with the DMPS and the APS. In the present work it was determined from the scattering measured with the nephelometer with the alternating PM1-PM10 inlet. In the present work we omitted the PM1/PM10 ratio calculation from Mie modeling, however, so the ratios are not strictly comparable. Both approaches have fairly large uncertainties associated with the large particles.

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

I have tried to stress this in text. But now there are also RFE calculated for more realistic conditions and ambient moisture.

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

Added the phrase.

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

Replaced.

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.

I added the citations here and included some discussion too.

"These relationships were also observed in a study of AOPs at the Station for Observing Regional Processes of the Earth System (SORPES), a measurement station in Nanjing China (Shen et al., 2018). Also, Delene and Ogren (2002) and Sherman et al. (2015) observed similar systematic variability between σ_{sca}, ω_0, b, and $RFE_{H\&S}$ at several North American measurement stations; when the σ_{sca} increases, the ω_0 increases and the b decreases. Sherman et al. (2015) suggested that this variability could be caused by deposition of larger particles, which typically absorb less light. Delene and Ogren (2002) observed that $RFE_{H\&S}$ increases (i.e. becomes less negative) with increasing σ_{sca}, but Sherman et al. (2015) did not observe this trend. "

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?

I added some discussion here. The larger accumulation mode particles could be aged pollution particles that have been grown by SO_2 for example. The emissions of SO_2have decreased, which would support this claim.

"The results, which are presented in Fig. S3, pointed out that relatively greatest decrease occurred for accumulation mode particles that were 500 – 800 nm in diameter. On average, the volume size distribution of accumulation mode particles peaks around 300 nm (see Figs. S3 and S4) so the greatest decrease occurred at the larger sizes of the accumulation mode. The decrease in this size range might be caused by decrease in long-range transported pollution. Aged pollution particles might be grown by other substances, such as SO_2 in the atmosphere so their sizes are larger than freshly emitted or formed particles. SO_2 emissions have decreased in Europe (Tørseth et al., 2012), which supports this assumption. A trajectory analysis by Virkkula et al. (2011) showed that α_{sca} was clearly higher in air masses from continental Europe than from the North Atlantic and but also that the highest α_{sca} values were measured in air masses sources from within southern Finland, which would suggest that larger particles are not from nearby the station."

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

I have modified the figure and also made the legends bigger.

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?

I fixed this. The whiskers were drew with dashed lines, which caused them to be not attached to the boxes like you noticed in your next comment.

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.

Fixed this by drawing the whiskers with solid lines.

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

Yes it should! I fixed the number.