

Replies to the reviewers' comments

The authors thank the reviewers for their evaluations. We believe answering the questions helped improving the paper. The text was corrected according to most of the suggestions of the reviewers. The largest changes were

1) In the section 1. Introduction we added text describing the geographical and temporal availability of data on aerosol optical properties, cloud condensation nuclei, and particle number size distributions. This was done as a reply to the comment of reviewer #2.

2) We added a description of two the backscatter fraction (BSF) and scattering Ångström exponent (SAE) as a new subsection **2.3 Optical properties calculated from the nephelometer data**. The section shows both the equations and a qualitative description of the relationship of particle size, BSF and SAE. We also added the section **2.6 Aerosol optical properties and CCN concentrations of simulated size distributions**. These additions were done in order to help discussions later in the paper.

3) In order to make the paper more clearly describing also aerosol physics, not only the parameterization, we changed the titles of the sections from

1. Introduction
2. Methods
 - 2.1 Sites and measurements
 - 2.2 Data processing
 - 2.3 Light scattering calculated from the particle number size distributions
 - 2.4 CCN number concentration calculated from the particle number size distribution
3. Overview of measured properties
 - 3.1 AOPs and CCN calculated from particle size distributions
 - 3.2 Relationships between AOPs and CCN
4. Development of the parameterization
 - 4.1 Site-dependent parameterization for each measured supersaturation, $N_{CCN}(AOP_1)$
 - 4.2 General combined parameterization $N_{CCN}(AOP_2)$
5. Results and discussion
 - 5.1 Comparison of N_{CCN} from the AOP parameterization and measurements
 - 5.2 Evaluation of the effect of particle size distribution to the parameterization
 - 5.3 Aerosol size characteristics for all sites
 - 5.3.1 Diurnal variation of particle number size distribution
 - 5.3.2 Distribution of geometric mean diameter
 - 5.3.3 Contribution of light scattering by sub- μ m particles
6. Conclusions

to

1. Introduction
2. Methods
 - 2.1 Sites and measurements
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 - 2.3 Optical properties calculated from the nephelometer data**
 - 2.4 Light scattering calculated from the particle number size distributions
 - 2.5 CCN number concentration calculated from the particle number size distribution
 - 2.6 Aerosol optical properties and CCN concentrations of simulated size distributions**
- 3. Relationships between N_{CCN} and AOPs**
 - 3.1 Site-dependent N_{CCN} - AOP relationships**
 - 3.2 Site-independent relationships between N_{CCN} , AOPs and supersaturations**
- 4. Analyses of size distribution effects on N_{CCN} - AOP relationships**
 - 4.1 N_{CCN} - AOP relationships of simulated particle size distributions**
 - 4.2 Aerosol size characteristics of the sites**
 - 4.2.1 Diurnal variation of particle number size distribution**
 - 4.2.2 Distributions of geometric mean diameters**
 - 4.3.3 Contribution of light scattering by sub- μ m particles**
- 5. Conclusions**

4) We included also more supplementary material in order to answer reviewers' questions.

5) There is a minor change also in the values of the parameterization. In the discussion paper it was

$$N_{CCN} \approx ((287 \pm 45)SAE \ln(SS/(0.093 \pm 0.006))(BSF - BSF_{min}) + (5.2 \pm 3.3))\sigma_{sp}$$

but in the revised version

$$N_{CCN} \approx ((286 \pm 46)SAE \ln(SS/(0.093 \pm 0.006))(BSF - BSF_{min}) + (5.2 \pm 3.3))\sigma_{sp},$$

The change is due to rechecks of the calculations. Since the final result - the above equation - is slightly different it is obvious that also some of the intermediate results are slightly different from those in the discussion paper.

6) We have added more atmospherically relevant and qualitative interpretations throughout the paper.

In the revised text major changes and additions are written using red letters. Minor language corrections have not been indicated.

Below the reviewers' questions are written in bold letters and the replies are intended.

Detailed replies to Anonymous Referee #1

Major and General Comments:

1) Page 1 line 26 of the Abstract is misleading. The manuscript focusses on the relationship between AOD and CCN, this parametrization also depends on the s,m upersaturation, however the manuscript does not show that the relationship between measured CCN number concentration and the corresponding supersaturation (S) is logarithmic, there is even no figure and no discussion about the relationship between CCN and S in the manuscript.

We don't quite understand the reviewer's comment because the derived formula

$$N_{CCN} \approx ((286 \pm 46)SAE \ln(SS/(0.093 \pm 0.006))(BSF - BSF_{min}) + (5.2 \pm 3.3))\sigma_{sp},$$

includes a logarithmic dependence on SS. The section where this was derived included discussion also. However, we have now included a new figure that shows this clearly. Fig. 1 of the revised paper shows the averages of the measured N_{CCN} at the six sites at the station-specific supersaturations of the CCN counters and a logarithmic fitting to the data. Also in the figures of section where the equation is derived the logarithmic and power function fittings are presented and discussed.

2) The manuscript is very technical, following the manuscript would have fit perfect in the journal aerosol measurement techniques as well. Please clarify why this is an ACP rather than an AMT manuscript.

Here is our argumentation for keeping the paper in ACP. In the paper we processed data from from six stations in very different environments around the world. We developed a parameterization to estimate the number concentration of cloud condensation nuclei (N_{CCN}) from measurements of aerosol optical properties. We have also studied how the dependence of N_{CCN} on supersaturation affects the relationships of N_{CCN} and AOPs. We first show that with the new Fig. 1 where N_{CCN} vs. SS is plotted for all the stations and a logarithmic function fits the data clearly better than a Twomey-style power function.

We found a formula that combines data from all six sites, it is approximately

$$N_{CCN} = ((286 * SAE * \ln(SS/0.093) * (BSF - constant) + 5.2)\sigma_{sp}$$

where SAE is the scattering Ångström exponent, BSF the backscatter fraction, σ_{sp} the total scattering coefficient and SS the supersaturation. This formula shows that N_{CCN} depends on SAE, BSF and σ_{sp} and that there is a logarithmic dependence on SS. If the data were fitted by using a power function of SS the fitting was not as good as by using a logarithm of SS. See the revised Figure 5.

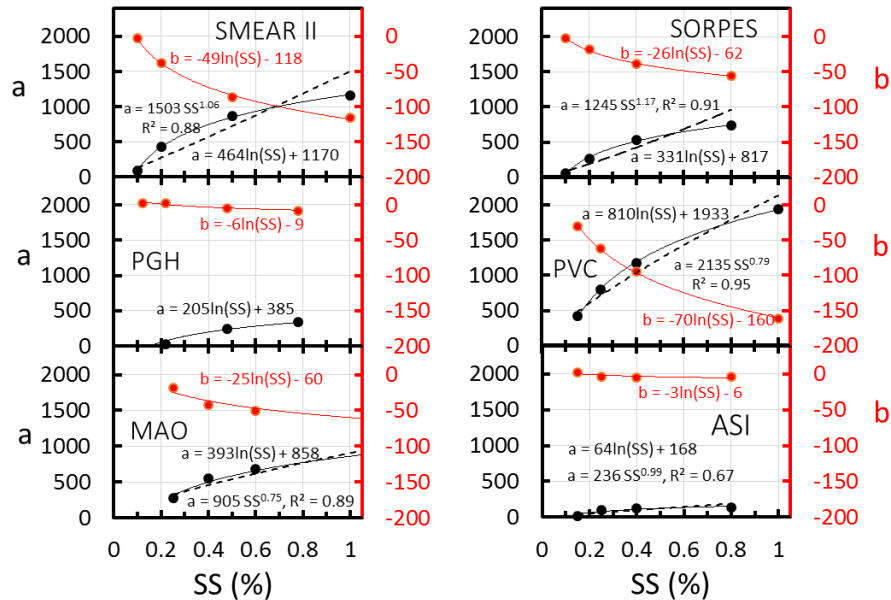


Figure 5. The coefficients a and b of each station (Table 2) as a function of supersaturation. Two types of functions, a logarithmic and a power function were fitted to the coefficient a, to coefficient b only a logarithmic function. The squared correlation coefficients R^2 are shown only for the power function fittings, for the logarithmic fittings they were all > 0.99 .

We simulated aerosol size distributions and obtained similar relationships between N_{CCN} and aerosol optical properties as in the field data. The above arguments show that the formula that we present can be considered either as only a parameterization or also as a representation of a physical phenomenon, not tied to a geographical location. This can be considered atmospheric physics.

In addition we studied the uncertainty of the method and the explanations of the uncertainties based on the aerosol size distributions in different environments. This is also atmospheric physics.

These are the main arguments we considered supporting the publication in ACP. We have added more atmospherically relevant and qualitative interpretations throughout the revised paper.

3) Table 1 it would be very helpful to add a small description of the site.

We have modified the table accordingly.

4) Page 6 line 14 the authors mention that no severe pollution episodes were observed in the data used for this study. However, table 1 shows that the data used for this study span a whole year. Was the data selected for no pollution periods and if so, what was the criterium?

The data were not purposely selected, only very few ($<<1\%$) suspicious data points were excluded. The percentage of excluded data is also added into the manuscript (supplement S1).

We also change the paragraph accordingly. he claim that no severe pollution episodes were observed is a qualitative, not a very precise statement. The paragraph was replaced by

MAO refers to Manacapuru in Amazonas, Brazil. It is a relatively clean site where Manaus pollution plumes and biomass burning plumes impact the background pristine rainforest aerosol alternately (e.g., de Sá et al., 2019).

5) Page 7 line 3-9 describes which data have been excluded for the analysis. It would be nice to mention the percentage of excluded data by the different criteriums and in total.

Detailed description and percentages of excluded data are presented in the supplement S1.

6) Page 9 line 9-11: The authors describe that NCCN was calculated by integrating the number concentration of aerosol particles larger than a certain diameter. How these diameters were chosen? Using the Köhler Theory and a certain hygroscopicity for the particles? If yes, which hygroscopicity have been used?

In the discussion paper we had chosen the critical diameters in the range of 80 - 110 nm intuitively for the simulation. There was no proper reasoning for this selection. When replying the reviewer's question we calculated that this corresponds to the supersaturation range of 0.213 – 0.344 when the global average hygroscopicity parameter kappa of 0.27 is used. This range is narrow so we repeated the simulations using a critical diameter range of 50 – 150 nm which is more reasonable that corresponds to a SS range of ~0.14 – 0.74% with the same kappa.

7) Page 10 line 28: The authors mention that the error in the measurements influence the result of the linear regression. As long as the linear relationship between measured variables which both have an uncertainty plays a key role in this study, the author should use a bivariate regression including the uncertainty of all measured variables as introduced by Cantrell 2008.

We have now repeated the calculations by using Reduced Major Axis (RMA) regression. The main results don't change although the derived function's constants are slightly different, see the [supplement S2](#).

8) Page 11 line 18: Is there also a lower R^2 for higher N_{CCN} concentrations? Figure 8 b suggest such a closure.

Yes, we agree with the reviewer, it shows clearly in figure 8(b) that the estimated N_{CCN} is more consistent with measurements at higher CCN concentrations. We split the whole data set into two groups, one is called 'lower 50%', corresponding to the time when $N_{CCN}(meas) < N_{CCN}(meas)(Median)$ while the rest is called 'upper 50%'. The SS for the split is set as a moderate SS% which is 0.3~0.4% depending on site. It is clear that at almost at all cases R^2 for 'upper 50%' groups are higher than 'lower 50%' groups. However, the subset has a narrower range of variation. The R^2 for 'upper 50%' groups are still lower than the R^2 of the whole data set. The details are shown in the table below.

		R^2		
		Whole	Lower 50%	Upper 50%
SMEAR II	0.10%	0.66	0.28	0.54
	0.20%	0.84	0.36	0.65
	0.50%	0.75	0.38	0.48
	1.00%	0.54	0.38	0.43
SORPES	0.10%	0.59	0.16	0.26
	0.20%	0.75	0.38	0.73
	0.40%	0.68	0.40	0.71
	0.80%	0.66	0.33	0.60
PGH	0.12%	0.81	0.48	0.73
	0.22%	0.83	0.57	0.72
	0.48%	0.79	0.63	0.61
	0.78%	0.74	0.57	0.55
PVC	0.15%	0.83	0.36	0.77
	0.25%	0.84	0.35	0.81
	0.40%	0.84	0.31	0.80
	1.00%	0.81	0.19	0.78
MAO	0.25%	0.81	0.32	0.79
	0.40%	0.83	0.36	0.80
	0.60%	0.74	0.30	0.65
	0.80%	0.69	0.22	0.59
ASI	0.15%	0.84	0.43	0.82
	0.25%	0.74	0.26	0.64
	0.40%	0.65	0.17	0.47
	0.80%	0.54	0.18	0.30

We do not add that into the text since the paper is already long enough. However, we added this brief discussion into the revised paper's section 3.2:

The correlation coefficient of $N_{CCN}(AOP_2)$ vs. $N_{CCN}(meas)$ is higher at higher CCN concentrations (not shown in the figure). One possible reason for this is that when CCN concentration is lower, the aerosol loading is usually lower and also the relative uncertainties of both N_{CCN} and AOPs are higher than at high concentrations.

9) Page 12 line 12: What is a reasonable long period?

Thank you for this question. We added an analysis in the supplement (S5. Analysis of the uncertainty related to the number of samples). It shows that when the number of hourly samples is > 1000 the uncertainty of the BSFmin is low enough.

10) Page 12 line 22-23: What is the percentual uncertainty?

Yes we agree it would be helpful to present the percentual uncertainty. We rewrote the paragraph as below:

Using 287- SAE_{10} overestimates or underestimates a_1 by +41%, +30%, -20%, -32%, -26% and +252% for SMEAR2, SORPES, PGH, PVC MAO and ASI, respectively. It brings additional uncertainty in the estimation and the uncertainty may be very large is a_1 is very small. Nevertheless SAE_{10} is the only parameter we found that positively related with a_1 and can easily been obtained from measurement at current stage. Searching for a more suitable proxy for a_1 would be an important part of follow up studies.

11) Page 14 line 6-7: The authors wants to show the different magnitude in Figure 9a, following I would recommend to use only one axis, by that the different magnitudes would be visible easier.

In the revised version the $R_{CCN/\sigma}$ of the wide size distributions are plotted twice: the black symbols and line use the left axis to emphasize the big difference in the magnitudes of the wide and narrow size distributions; the red symbols and line use the right axis to show that the shape of the $R_{CCN/\sigma}$ size distribution is very similar to the one calculated for the narrow size distributions.

12) Page 16 line 24: The MAO site was measuring aerosol downwind of the City Manaus and not Amazon rainforest air. The pronounced nucleation size particles suggest predominant anthropogenic emissions from Manaus.

We thank the referee for pointing this out. We modified the manuscript accordingly as below:

NPF is rarely observed in the Amazon forest as reported by Wang et al. (2016). However, it does take place at MAO as is shown in the diurnal cycle of PNSD. The reason is probably that the MAO site was measuring aerosol downwind of the City Manaus. At ASI, there no evidence of NPF according to the PNSD diurnal cycle.

13) Page 17 line 14: The authors compares the result from MAO with the results from ATTO. The disagreement of this comparison is a strong indication that the air masses measured at MAO are not representative for the Amazon rainforest. Please clarify this aspect.

We thank the referee for pointing this out. It was a mistake to make a comparison between ATTO and MAO. Now we clearly realize that even though ATTO and MAO are only ~100km from each other they represent different location and air masses. We removed the comparison from the manuscript.

Minor Comments

- The authors always write 'SS%' instead of 'SS' or 'S' for the supersaturation. The '%' behind the value is sometimes used, sometimes not. I would recommend to use 'SS' or 'S' for the supersaturation and always mention the unit behind a variable.

We have changed all SS% symbols to SS according to the reviewer's suggestion

- Page 13 line 5: two times "of"
corrected

- Page 14 line 9: is this a complete sentence?

No. The sentences " Note also that the rates of decrease of $R_{CCN/\sigma}$ and BSF. We used this information for estimating particle sizes with a stepwise linear regression." were replaced by

"The decrease of $R_{CCN/\sigma}$ and BSF with growing GMD was used for estimating particle sizes with a stepwise linear regression."

- Page 16 line 1: is the double negation on purpose?

No, it was a typing error. Corrected.

Detailed replies to Anonymous Referee #2

The paper is on an interesting subject and the parameterization of CCN concentrations by optical parameters can be useful. The data are carefully analyzed and the parameterization is developed step by step, which helps understanding the resulting complicated formula. Overall, I find that this is a paper that is in principle publishable after moderate to major revisions. One reservation I have, is that it is clearly a method paper that can certainly be very useful, but does not have major scientific results (as to e.g., actual CCN in various environments and larger implication), so ACP is in my opinion not the best journal for this. It fits much better in a method-oriented journal. I would strongly suggest to submit it somewhere else.

Reviewer 1 asked the same question. See the reply 2) above.

Parts of the steps taken could be better motivated and better interpreted (see comments below), which would make the manuscript easier to follow and less technical and dry. Overall I think the length of the paper and the 13! Figures are a bit excessive. Please think of a way to summarize the information in a more compact way.

We have revised the text to make it a bit less technical. This was done by changing the titles of the sections to more descriptive ones and by adding more interpretations related to size distributions at the different sites. We removed Fig 3. of the discussion paper but in order to answer reviewer #1's question 1) about showing the logarithmic dependence of N_{CCN} on SS we added a new Fig 1. The number of figures is still 13. There are several papers also in ACP that have 13 figures so we don't consider that to be too many.

The writing needs to be improved overall and the English corrected in some places (many formulations are unclear and sometimes grammatically incorrect).

We have now corrected many parts of the paper.

Specific comments:

1) Page 3, line 4: Explain better, why they are not adequate. I assume you mean the spatial coverage? Are optical properties really more widely measured than size distributions? Support this claim!

Yes, the spatial coverage of particle size distribution data is indeed clearly smaller than that of the optical properties. The WMO Global Atmosphere Watch World Data Centre for Aerosols (GAW-WDCA) (<http://ebas.nilu.no/>) contains on 20 June 2019 particle number size distribution data sets from 22 countries altogether from 58 stations, but only 5 are outside of Europe. The CCNC data are from 3 European sites. Light scattering coefficients measured with a nephelometer are from 31 countries and 103 stations that are located on all continents and also on some islands. The temporal coverage data in the GAW-WDCA data base is such that the oldest nephelometer data, those from Mauna Loa start in 1974 whereas the oldest particle number size distribution data, those from SMEAR II start in 1993. Another easily available source for data is the US Department of Energy Atmospheric Radiation Measurement (ARM) user facility (<https://www.arm.gov/data>). On 20 June 2019 we found that the ARM research facility data contains particle size distribution data from 7 permanent sites and light scattering coefficient measured with a nephelometer at 20 sites. It is clear that there are other data sets of all of these measured around the world but those that can be found either from the GAW-WDCA or the ARM data bases are quality controlled and readily available.

2) Page 3, line 26: Do you mean N_{CCN}/σ_{sp} instead of R_{CCN}/σ_{sp} ? Later you call this ratio $R_{(CCN/\sigma)}$ please be consistent.

This is a good observation, we have corrected this.

3) Page 8, line 6-9: This paragraph consists of several incomplete, grammatically incorrect sentences and as a result, it is unclear what you are actually doing in this calculation. Please formulate clearly and explain in more detail.

Corrected.

4) Page 9, line 5: How well do you think a unimodal size distribution can describe a realistic situation, where size distributions are at least bi-modal? Give some motivation already here for choosing this approach.

The following paragraph was added:

Using a unimodal size distribution for the simulation is an approximation only. In the boundary layer particle number size distributions consist typically of an Aitken mode in the size range $\sim 25 - 100$ nm, an accumulation mode in the size range $100 - 500$ nm, and during new particle formation also a nucleation mode in the size range < 25 nm in very different environments (e.g., Dal Maso et al., 2005; Herrmann et al., 2015; Qi et al., 2015). While the total number concentration is dominated by the smaller modes, essentially all light scattering is due to the accumulation mode and also coarse particles in the range of $1 - 10$ μm . For instance at SMEAR II the average contribution of particles smaller than 100 nm to total scattering was ~ 0.2 % and even at the end of new particle formation events not more than ~ 2 % (Virkkula et al., 2011). On the other hand, also most of the CCN are also in accumulation mode range, especially for low supersaturations ($SS < 0.2\%$), at higher SS also in the Aitken mode (Sihto et al., 2011).

5) Page 9, line 13: The section is called “Overview of measured properties”, but it does not contain an overview of measured properties, more a quality control. It should be merged with the method section.

We agree, as it is in the discussion paper the section really is not an overview of the data. But now we have rewritten the beginning of section 3. First, the titles were changed from

- 3. Overview of measured properties
- 3.1 AOPs and CCN calculated from particle size distributions
- 3.2 Relationships between AOPs and CCN

to

- 3. Relationships between N_{CCN} and AOPs
- 3.1 Site-dependent N_{CCN} - AOP relationships
- 3.2 Site-independent relationships between N_{CCN} , AOPs and supersaturations

Second, an overview of the data at the different sites is given in the new section 3.1.

6) Page 9, line 14-24: This Paragraph seems out of place here. At the beginning of a new section, before the sub-sections, I would expect some general introduction. Instead we get some details of only one station that are neither well explained nor well motivated. Please rearrange this section and find a better place for this information (after you clarify it).

We agree and have modified the paragraph and the section 3. We have added a short introduction at the beginning of the sections.

7) Page 9, line 19: You do not make clear how you make this fit (e.g., how you determine k) and why it does not fit. Is this relevant for the paper in any way? It well known that an error function gives a much better fit to CCN spectra.

The reviewer is right, this is not relevant. We removed this. We also rewrote the beginning of section 3.

8) Page 9, line 24: From the previous discussion it is not clear, why/how this parameterization involved absorption data. If this paragraph is absolutely necessary, please explain in more detail, otherwise I would suggest to omit this discussion entirely.

The reviewer is right, this is not relevant. We removed this.

9) Page 10, line 13/14: Give an interpretation of this fact and a motivation, why you investigate this dependence.

The interpretation is that when BSF is constant the shape of the particle size distribution is also very probably constant in the particle size range significantly affecting light scattering. Then the increase of scattering coefficient is very probably due to the increase in the particle number concentration in the same size range and then also the number of CCN. Fig. 1 alone does not show how NCCN depends on BSF so it serves as a motivation of the continuation to the use of Eq. (3) and the subsequent analyses.

10) Page 11, line 11/12: Please use the appropriate reduced major axis regression then, instead of a regression that is not suitable for your data

We have now repeated the calculations by using Reduced Major Axis (RMA) regression. The main results don't change although the derived function's constants are slightly different, see the supplement S2.

11) Page 12, line 3/4: I am not sure what you mean by "underlying reason"? b_0 is the limiting value for $SS=1$ and the range of SS stays the same. So go from N_{CCN} 0 at approx. SS 0 to a higher b_0 at SS 1, obviously $\ln(SS)$ must be multiplied by a higher b_1 . I would omit the corresponding figure 6.

The scatter plot in Fig. 6 is probably the most important one in the paper since it shows that the constants are linearly dependent on each other. The linear relationships are then used to reduce free parameters. The regressions yield the constants needed for the development of the general parameterization, Eq. (6). We do not omit Fig. 6.

It is true that b_0 is the limiting value for $SS = 1\%$ but the absolute values of the constants are by no means obtained from the data without the regressions. First, not all the stations even have measurements at $SS = 1\%$ so there is no way to deduce the values of these constants without the fitting to the data. Second, it is by no means clear that the constants of all the stations would follow straight lines. For instance, why would the pairs (b_1, a_1) of all the stations be distributed approximately along straight lines? But plotting them to the same figure they do. Why wouldn't the forms of the scatter plots be nonlinear? Or even random? There is no a priori reason. This is what we try to study by the simulations. And that is what we are wondering by writing about an "underlying reason". But nevertheless, we remove this expression.

12) Page 12, line 19: Why? try to give a qualitative interpretation of this fact. . .

The coefficient a_1 is positively correlated with SAE which means that the higher SAE is the higher is N_{CCN} at a constant σ_{sp} . Light scattering and its wavelength dependency is highly dependent on particle size so SAE is often used as an indicator of the particle size distribution. It is often inversely related to particle size: at $SAE > 2$, the volume size distribution is typically dominated by particles smaller than $0.5 \mu m$, and at $SAE < 1$, larger particles ($D_p > 0.5 \mu m$) dominate the distribution. So, the positive correlation of SAE and a_1 means that when volume size distribution is dominated by small particles the CCN concentration is high. We have added several qualitative interpretations into the text.

13) Page 14, line 13ff (Fig10): I would need a better motivation here, why these piecewise linear fits were tried in the first place – looking at this figure an exponential fit would be much more appropriate and at this point it is not clear what the authors are trying to do here – it gets a bit clearer later on, but a good explanation and motivation here would help. What does the linear approximation represent physically?

It is true that an exponential fit would be perfect to explain the relationship between $R_{CCN/\sigma}$ and BSF. But this is not at all what we are looking for. We are looking for the slopes and offsets in the relationship $R_{CCN/\sigma} = aBSF + b$ that was used for fitting the field measurement data. So, physically it would mean that N_{CCN} would increase linearly as a function of BSF even though this is not exactly correct.

We assume that the simulated data consists of a series of lognormal size distributions. This would be simulating some station data where the GMD of size distributions varies in a certain range. Then application of linear regression yields the slopes and offsets and we can find out how these are related to each other. As it proves out, the simulated b vs a pairs follow the lines that are similar to those obtained by plotting the slopes and offsets of the actual field data (Fig. 12).

14) Page 14, line 16: CMDe has not been clearly defined here at first use – what is a “particle size that is used for describing the size range of each regression”?

On lines 16 – 18 it is written " ... we define here as the equivalent geometric mean diameter GMD_e , the geometric mean of the range of the GMDs of the unimodal size distributions used for each regression." Geometric mean of the range of the GMDs is the square root of the product of smallest GMD and the largest GMD of the range. We do not understand how this could be more clearly defined. It can also be presented with symbols as $GMD_e = \sqrt{GMD_{low} GMD_{high}}$, where GMD_{low} and GMD_{high} are the smallest GMD and the largest GMD of the range, respectively. But it is probably clarified with examples. Above, on line 12 two examples were given of the regressions, one calculated for the GMD range from 50 nm to 100 nm and the other for the GMD range from 100 nm to 200 nm. The GMD_e s of these two size ranges are 70.7 nm and 141.4 nm, respectively.

15) Page 14, line 27: In line 22 you described the different behavior of slope and offset here you say that they vary simultaneously? Please be precise with the language. Also “simultaneously” implies temporal variation, which is not the case here . . .

We removed the sentence "However, they decrease and increase simultaneously." and modified the corresponding paragraph. It was replaced by

Note also that the ranges of the absolute values of the slopes and offsets of the narrow and wide size distributions are very different. For instance, when $GMD_e = 100$ nm the slope $a \approx 4000$ cm^{-3}/Mm^{-1} and $a \approx 1600$ cm^{-3}/Mm^{-1} for the narrow and wide size distribution, respectively. Since $N_{CCN} = R_{CCN/\sigma} \cdot \sigma_{sp} = (aBSF + b)\sigma_{sp}$ this means that the N_{CCN} of narrow size distributions is more sensitive to variations in mean particle size than the N_{CCN} of wide size distributions.

16) Page 14, line 27/28: “This is the link . . .” What precisely is the link? You discussed several variations in the sentences before, but none of them seem a clear “link” to me.

We removed the sentence " This is the link to the observations from the field stations." and rewrote the corresponding paragraph. See the answer to the referee comment 15 above.

17) Page 15, line 9-13: This argument is very technical and also in my opinion the linear relationship has nothing to do with dependence on GMD and GSD (see point 12). There are much better, more qualitative arguments, why these coefficients should depend on the size distribution and how – please argue more along those lines.

The reviewer is right. Our argument was wrong, the simulations did not show the linear relationships between the coefficients a_0 , a_1 , b_0 and b_1 that are the factors from the fittings of $(a_1 \ln(SS) + a_0)BSF + b_1 \ln(SS) + b_0$ with the data. The simulations yielded only the slope a and the offset b of $aBSF + b$ but not their dependence on SS . To obtain the factors a_0 , a_1 , b_0 and b_1 we should have added also a simulation as a function of SS . That was not done. In order not to add new figures, we did not include a figure where the simulated a vs SS and b vs SS is shown.

However, the simulation clearly shows that both the slope a and the offset b depend very strongly on both the geometric mean diameter (GMD) and the geometric standard deviation (GSD) of the size distribution (Fig. 11). We do not see any other way but the simulation to show the dependence of a and b on the size distribution.

We have replaced the corresponding paragraph by

Several observations can be made of Fig. 11. First, for the simulated wide size distributions the relationship of the offset and slope is unambiguous but not for the narrow size distributions at sizes $GMD_e > \sim 200$ nm (Fig 11b). Secondly, the field data points roughly follow the lines of the simulations. This suggests that the slopes and offsets of the linear regressions of $R_{CCN/\sigma}$ vs. BSF yield information on the dominating particle sizes just as they do for the simulated size distributions. For instance, the PVC data point corresponding to the highest supersaturation has the highest slope ($1970 \text{ cm}^{-3}/\text{Mm}^{-3}$, Table 3) and it is close to the wide size distribution line with the activation diameter of 50 nm (Fig. 11a). This corresponds to the GMD_e of ~ 150 nm (Fig. 11b). The SMEAR II high SS offset vs. slope fits best with the corresponding lines of the narrow unimodal size distributions with activation diameters in the range of $\sim 50 - 110$ nm and the corresponding $GMD_e \approx 150 - 200$ nm.

18) Page 18, line 17/18: Please give an interpretation of this fact.

The reviewer refers to this sentence: " However, a comparison of the correlation coefficients and the $\sigma_{sp}(PM_{11})/\sigma_{sp}(PM_{10})$ frequency distributions of each site shows the opposite. " and to the list of sites in an order of decreasing correlation coefficients in the next sentence.

This means that the correlation coefficient of N_{CCN} calculated from the aerosol optical properties ($N_{CCN}(AOP_2)$) vs. the N_{CCN} measured directly ($N_{CCN}(meas)$) is the higher the larger fraction of total scattering is due to large, PM_{10} aerosol. This suggests that N_{CCN} can be estimated better from the aerosol optical properties for sites dominated by large particles than for sites dominated by small particles. This further suggests that the ambient size distributions were so wide that the nonmonotonous relationship between particle size and BSF discussed above did not play an important role.

19) Page 19, line 15/16: What coefficients are you taking about? There are many discussed in the manuscript. Please be more precise with your language throughout the manuscript.

The sentence is about the coefficients a_0 , a_1 , b_0 and b_1 in the formula

$$N_{CCN}(AOP_2) = (aBSF + b)\sigma_{sp} = (a_1\ln(SS)+a_0)BSF + b_1\ln(SS)+ b_0)\sigma_{sp}.$$

However, we removed that sentence from the conclusions of the revised paper. The conclusions now contain more atmospherically relevant and qualitative interpretations of the analysis.

Minor comments:

Page 1, line 27: SAE₁₀ and BSF_{min} not defined

We replaced SAE_{10} simply by SAE since it was just in the previous sentence written that the parameterization deals with PM_{10} particles. BSF_{min} has now been defined.

Page 2, line 1: supplementary -> replacement (or similar), "supplementary" means supporting or additional; please check for similar mistakes throughout the manuscript, or have it proofread by a native speaker

The sentence

This makes particle number size distribution measurements capable of serving as a supplementary of direct CCN measurements.

was replaced by

This makes particle number size distribution measurements capable of substituting direct CCN measurements.

Page 2, line 4: Do you mean "or" instead of "of", otherwise this sentence does not make sense to me.

Yes, "of" was a typing error. Corrected.

Page 4, line 25: What do you mean by “likewise in Smale . . .”?

"likewise in" was replaced by "similar to"

Page 5, line 28; “Instrument mentors” is not a valid term, please check manuscript throughout for such wrong use of vocabulary.

“Instrument mentors” were removed and the sentence

The supersaturations are typically calibrated before and after each campaign at an altitude similar to measurement site by instrument mentors according to CCN handbook (Uin, 2016b).

was replaced by

The supersaturations are typically calibrated before and after each campaign at an altitude similar to measurement site according to the CCN handbook (Uin, 2016b).

Page 6, line 5: coal is also a fuel

" coal and fuel combustion " was replaced by " coal combustion "

Page 6, line 9: is this not a coastal site, rather than marine?

Yes, Cape Cod is a coastal site, not marine. Replaced.

Page 6, line 9: “emphase” is not a word

The sentence where the word "emphase" was is

"Ascension Island (ASI) locates in the southeast Atlantic where westward transport of southern Africa biomass-burning aerosols emphases heavy aerosol loading."

It was replaced by

"Ascension Island (ASI) is located in the southeast Atlantic where westward transport of biomass-burning aerosols from southern Africa may increase aerosol concentrations to high levels."

Page 6, line 9/10: The sentence is not grammatically correct

The paragraph on ASI was corrected.

Page 16, line 13: is -> are

Corrected