Response to Reviewer 1:

The authors would like to thank Anonymous Referee #1 who helped us improve our manuscript.

According to the comments, we reconsidered and rectified our manuscript (ms). Below, we give a point to point reply. For convenience and to avoid an unnecessary inflation of this response letter, all corresponding changes in our revised ms are accordingly indicated. We refrain from listing all revised marginal fragments here. However, we present all essentially revised parts of the ms straight below our response *("in quotation marks and in italics").*

reviewer comment: The paper could be strengthened by including some discussion of the implications of this work towards climate change estimates, which is one of their stated motivations for the study.

answer: Our derived refractive index values refer to dry aerosol particles, as they are derived from dry aerosol number size distribution measurements. Unfortunately, we are not able to extend the discussion towards climate change estimates, because we have no sufficient information on the water uptake (hygroscopicity) of the aerosol particles from our measurement site yet. Therefore, we could only speculate on the real ambient light scattering and absorption coefficients, which we do not want. We hope that the planned hygroscopicity measurements this austral summer (2019-2020) at our measurement site will allow us to make such a study.

reviewer comment: There were several typographical errors in the text that I did not attempt to correct; I suggest another careful editing of the manuscript.

answer: A careful editing and inspection by an native speaker was performed.

reviewer comment: Page 1, Line 1-3: This sentence is a bit unclear. I think the authors are trying to state that reducing uncertainties in modeling evaluations of climate change require more accurate aerosol optical properties. It might also help to point out why it is important to have measurements of refractive index at the poles- is this because climate change is enhanced there relative to other locations? It would also help to state when these measurements were made earlier in the abstract.

answer: The sentence was rewritten, now it reads:

"Though the environmental conditions of the Weddell Sea region and Dronning Maud Land (DML) are still relatively stable compared to the fast-changing Antarctic Peninsula, we may suspect pronounced effects of global climate change for the near future (Thompson et al. 2011). Reducing the uncertainties in climate change modeling requires inter alia a better understanding of the aerosol optical properties, and for this, we need accurate data on the aerosol refractive index (RI). Due to the remoteness of Antarctica only very few RI data are available from this region (Hogan et al. 1979, Virkkula et al. 2006, Shepherd et al. 2018).

reviewer comment: Page 1, line 7: Do the authors refer to 2439 individual size distributions when they refer to measurement points?

answer: We referred to 2439 averaged size distributions, but it was removed now from the manuscript.

reviewer comment: Page 1, line 7: It is always helpful to also include wavelength and relative humidity conditions associated with optical property measurements. Sometimes people only read abstracts and figures.

answer: The wavelength and the RH condition are added into the abstract.

reviewer comment: Page 1, line 8: It would also help to include some uncertainty estimates or an estimate of standard deviation with the reported average RI_{eff}.

answer: The standard deviation value of RI_{eff} was added to the abstract.

reviewer comment: Page 1, line 13-19: This paragraph is a bit unclear. I understood it better after I had read the paper, but as part of the abstract it could benefit from clarification. Part of the issue is that the location of the site has not been described yet, so understanding the wind direction in respect to the site location and the Neumayer station is a bit confusing. It might be helpful to state the wind direction impact in a more generalized way, or provide more description of the site location first.

answer: We revised this paragraph:

"We find no significant dependence of the derived RI_{eff} values on the wind direction. Thus, we conclude that RI_{eff} is largely independent on the general weather situation, roughly classified in (i) advection of marine boundary layer air masses during easterly winds caused by passing cyclones in contrast to (ii) air mass transport from continental Antarctica under southern katabatic winds. Neumayer, the only relevant contamination source, is located 1.5 km north of the air chemistry observatory, where the measurements were performed. Given that northerly winds are almost absent, the potential impact of local contamination is minimized in general. Indeed our data show no impact of local contamination on RI_{eff}. Just in one case, a temporary high contamination episode with diesel engines operating right next to the measurement site resulted in an unusual high RI_{eff} of 1.59, probably caused by the high black carbon content of the exhaust fumes."

reviewer comment: Page 1, line 20: Referring to the time-averaging here is also confusing without having read the paper first. It may not be necessary to include in the abstract. Are these differences larger than the uncertainties of the measurements?

answer: We now remove this confusing sentence from the abstract.

reviewer comment: Page 1, line 20: It would also help here to state something about the larger implications of this work, trying back to the point of the study so that the reader grasps the larger importance of the work.

answer: To this end we added the following text to the abstract:

"To conclude, our study revealed largely constant RI_{eff} values throughout the year without any sign of seasonality. Therefore, it seems reasonable to use a single, constant RI_{eff} value of 1.44 for modeling optical properties of natural, coastal Antarctic sub-µm aerosol."

reviewer comment: Page 2, line 14: Probably the most common method would be from volume-weighted calculations of composition data because these are generally more available than detailed size distribution and scattering measurements.

answer: The reviewer is right, "The most common method" was changed to "A common method" *reviewer comment:* Page 2, line 24: It is important to point out that these methods do not directly measure refractive index- they use closure studies between a variety of measurements to determine the refractive index that provides agreement.

answer: The following sentence was added to the manuscript:

"We have to keep in mind that all above mentioned methods are not direct measurements of the RI. All of these methods search for RI values that provide good agreement in a closure study between different measured quantities."

reviewer comment: Page 2, line 25: Remove “parallel their” for clarity.

answer: We removed this term in the revised manuscript.

reviewer comment: Page 2, line 34: What does it mean, “Until the particles disappeared”?

answer: We changed the text to:

"until they could not see the particles in the microscope (i.e. until the applied oil's RI matched the RI of the collected particles)."

reviewer comment: Page 3, line 5: To what RH do these values correspond?

answer: All these values refer to dry aerosol, thus we modified the text to:

"In this paper we would like to present continuous data on the real RI at 633nm wavelength of the dry ambient aerosol as derived..."

reviewer comment: Page 3, line 5: It would also be helpful here to point out the importance of this work- why is it important to have yearlong estimates of RI from the Antarctic? What are the larger implications?

answer: The following text was added to clear this point:

“With this, our study aims at better understanding of the aerosol optical properties at a place where only very few such data are available with special focus on its temporal variability. Given the distinct seasonality of the aerosol composition (see Weller et al., 2008, Figs. 4 and 5 therein), we may likewise expect a seasonality of RI. To this end, continuous year-round data of RI are necessary, in particular regarding the lack of such measurements for the Antarctic realm.”

reviewer comment: Page 3, line 10: Include the study time period earlier, it will help when considering the information provided in the next paragraph.

answer: We modified this sentence to:

"The measurements presented in this paper were performed in the Air Chemistry Observatory (SPUSO from ”Spurenstoffobservatorium”) of the German Antarctic station of Neumayer III between February 2017 and January 2018."

reviewer comment: Page 3, line 31: Is RH and temperature measured? Define RH if it has not already been done.

answer: The RH of the aerosol entering the instruments was calculated from the measured outdoor temperature, RH and the measured indoor temperature. To clarify this point, we added a definition for RH and the following sentence to the text:

"The meteorological data used in this study (temperature, wind direction and speed and ambient RH) was measured directly on the roof of SPUSO."

reviewer comment: Page 5, line 8: How was 'penetration efficiency' determined?

answer: It is the ratio between the measured number size distribution with and without the tubing which causes the losses. We added the explanation to the text and renamed it to particle transmission efficiency.

reviewer comment: Page 6: Was there a quantitative measure by which "too noisy" was defined?

answer: We did not have a real quantitative measure, just the fact that no successful fit was possible, which became possible with further averaging.

reviewer comment: Page 6, line 7-8: This sentence is confusing, I suggest rewording.

answer: We reworded this confusing sentence and now it reads:

"This strong seasonal variability is the reason why in summer a much shorter time averaging period is sufficient to enable a successful RI_{eff} *fit. To keep the highest possible time resolution of the derived RI, we have chosen the length of the time averaging to be time dependent. And this length was determined by the actual particle concentration."*

reviewer comment: Page 6, line 10: How was "good enough signal to noise ratio" defined?

answer: Such that a meaningful RI_{eff} fit was possible. We now added this note to the text.

reviewer comment: Page 11, line 10: How long was the averaging time? I am not sure why rapidly changing aerosol should results in poor fit if both instruments are sampling the same aerosol at the same time? Was the aerosol changing faster than the SMPS could sample it? Did CPC data indicate this?

answer: The averaging time was variable, one hour at minimum, but up to 14 hours occasionally. Anyway, this is not the decisive point. The problem is if the aerosol changes significantly within one scan, which took 6 minutes for each instrument. The two instruments measure the number size distribution differently. The LAS captures all of the particles within this 6-minutes period, and therefore delivers an average number size distribution for this period. The SMPS on the other hand scans through the different diameters during 6 minutes and therefore captures only one size range/bin at a given time interval. More precisely: if for instance during the 6 minutes of a single scan the aerosol concentration increases appreciably but the shape of the distribution remains constant, the LAS returns the average number size distribution. In contrast, the SMPS returns a skewed number size distribution with too low concentrations in the lower, but too high concentrations in the upper size range (assuming that the SMPS is operated in an up-scan mode). We could also observe in the CPC data, when during the beginning of the scan the concentration was significantly different compared to the end of the scan. Accordingly, we changed the text to:

"The reason why the fit did not work in this case was that the aerosol population was significantly changing within the duration of the SMPS scan. During the first half of the scan an aerosol plume with very high concentration reached the instruments. This appears in the SMPS scan as a very high fraction

of small particles, because during the first half of the scan, the SMPS selected and measured the smaller particles. Contrary, the LAS captures all particles with different diameters at the same time, and therefore this event appears as an elevated overall concentration. This was an extreme and exceptional situation where some unavoidable construction was done around the SPUSO using machines powered by diesel engines."

reviewer comment: Page 11, line 20: Based on the stated estimated uncertainty (~3%), reporting this many significant digits seems unnecessary. This comment also holds for Table 1 and reporting of values throughout the paper. It would also help to report standard deviation for each month in Table 1

answer: We removed insignificant digits and supplied the corresponding standard deviation in the revised Table 1.

reviewer comment: Page 12, line 11: How was "significantly different from the yearly mean" determined?

*answer:*We determined the significance by a statistical T-test with a significance level of 0.01. The revised text now reads:

"These values are significantly different from the yearly mean (determined by using a statistical T-test with a significance level of 0.01)."

reviewer comment: Page 11, line 15: Providing standard deviations would help in discussing the lack of scatter in the data.

Answer: We added standard deviations in the revised text.

reviewer comment: Page 13, line 4-5: Organic carbon was not measured, can the authors comment on the possible contribution to mass at the site? Has it been measured during previous studies?

answer: Water soluble organic carbon (WSOC) was measured on filter samples during the austral summer in 2011 at our measurement site. This analysis revealed that the WSOC mass fraction (except methane sulphonate!) was on average less than 3% of the total mass and therefore we believe that excluding organic carbon from the chemistry based calculation does not influence significantly the results. We added this finding in the revised version:

"We do not have any information on the organic carbon mass fraction for our measurement period, and therefore we could not include this component into the calculation. However, previous water soluble organic carbon (WSOC) mass concentration measurements (Weller et al. 2015) showed, that in the austral summer of 2011 the WSOC average mass fraction was less than 3% and therefore we believe that organic carbon does not have a significant influence on the resulting RI. "

reviewer comment: Page 13: line 14: Typically, thermodynamics favor the formation of ammonium sulfate before ammonium nitrate, such that if there is enough ammonium available, it will neutralize sulfate before nitrate. What is the molar ratio of NH4/SO4 during the study? Were the aerosol acidic?

answer: The reviewer's objection is correct. The nss-SO_4^{2-}/NH_4^+ ratio is 11.2±8 (annual mean ± Stdev; the summer ratio is significantly higher) and we can generally assume acidic aerosol (at least during summer). Thus formation of ammoniumsulphate ($(NH_4)_2SO_4$) is more plausible. However, given the high nss-SO_4^{2-} excess (corresponding to a high H_2SO_4 excess), partly ammoniumbisulphate (NH_4HSO_4) may also be formed. The RI of NH_4HSO_4 is lower (Chýlek et al., 1995) than that of NH_4NO_3 (1.473 instead of 1.52), but the latter would be comparable to that of $(NH_4)_2SO_4$ (1.53; Tang, 1996). Unfortunately, in our case such a detailed chemical characterization is not possible, by neither our bulk aerosol nor our size segregated aerosol measurements. We considered this in the revised version of the manuscript (see revised Section 3.5).

reviewer comment: Page 14, line 15: The RIeff values look somewhat lower with East winds. It would help to expand the scale in Figure 6 to values ranging from 1.35-1.55 to sees these differences more clearly. Are these differences greater than uncertainty in the values themselves?

answer: We expanded the scale appropriately. Using the same significance test as used before showed us that the mean values are not significantly different from the yearly mean.

reviewer comment: Page 15. Line 3: The values in Figure 6 refer to an average value of all RIeff from the North. Did any individual distributions suggest contamination?

answer: No obvious contamination could be identified among these scans. If contamination was recognized (such as the scan in Figure 8), we removed those data from further evaluation.

reviewer comment: Page 16, line 12: Please provide units for scattering coefficient and a more detailed description of Cs. It must include a diameter-squared parameter.

answer: We provided units in the revised version. The scattering cross section has a diameter-squared unit, as it is now included in the text as well. The authors do not know what detail about Cs should be included additionally, because Cs is a well-known quantity. However, we mentioned the calculation method now in the text as well (i.e. a custom written Mie Code).

reviewer comment: Page 16, line 14. Is the scattering distribution in Figure 9 an average? Is the bimodal distribution is a function of the averaging of several different monomodal distributions at different times (suggesting interesting changes in the aerosol size distribution or refractive index).

answer: Yes, it is an average value, see line 14: "Figure 9 shows the time average of dσ/dlogD as function of the particle diameter". Both modes of the bimodal distributions (of both scattering coefficient and the number size distribution) are most of the time present simultaneously. In addition, we added to the main text:

“Considering the time evolution and not temporal averages we see, that these two peaks, as well as the two main peaks of the scattering coefficient size distribution, are often present simultaneously. In conclusion, the bimodality is not the product of time averaging of single modes appearing at different times.”

reviewer comment: Page 17: Figure 9 caption: What time period do these data correspond to? Is this an average of several distributions?

answer: Yes, it is an average of several distributions which is now stated in the figure caption.

reviewer comment: Page 17, line 9: This sentence is unclear

answer: Accordingly we changed the confusing sentence to:

"Moreover, the overall size distribution range can now be divided into 4 subranges suitable for separate RIeff calculations, representative for the corresponding subrange (Fig 10.)."

reviewer comment: Page 17, line 15-18: As Figure 2 shows, the instrument response at higher sizes shows a cross over region such that the instrument is unable to distinguish between refractive indices. Others have also shown this behavior at larger sizes (Garvey and Pinnick, 1983; Hand et al., 2000). Reporting refractive indices in these larger size ranges is probably not meaningful.

answer: Yes, the reviewer is right about the cross over region. As it can be seen in Figure 2, the m=1.4 and m=1.59 traces meet around log(D)=3 which is D=1000 nm. Our widest diameter range for size dependency investigations was 478-710 nm. Even at the largest diameter of 710 nm (log(710nm)=2.85) the two example instrument response curves are still very well separated from each other (almost at the orange arrow in Figure 2). Therefore, we think that the RI calculation is meaningful even at our highest diameter range. As the request of Reviewer #2, we removed this highest size range anyhow for another reason.

reviewer comment: Page 18, line 15: I am not sure what the authors mean by “geographic borders of this value’s validity”?

answer: We used the admittedly unclear notion “geographic border” to implicitly formulate the question: Where exactly can a single (seasonal independent) RI value of 1.44 be used? Is it valid only for our measurement site or also for other coastal or even continental Antarctic sites? We clarified this passage now to:

"Based on this, we recommend this single, temporally constant refractive index value for modeling of aerosol optical properties. In this context we suggest supporting investigations to examine the validity of this approach and the usage of seasonal independent RI_{eff} *values for the Antarctic region."*

reviewer comment: Page 18, line 14. Including experimental uncertainties here would help, as would restating the RI derived from composition data. Deriving Reff and the ability to calculate scattering coefficients using it and the measured size distributions, without the additional effort and cost of composition measurements, is an important benefit to this analysis. Comparing the RI derived from composition in the context of experimental uncertainty can strengthen the arguments for the importance of this type of analysis. In addition, composition measurements are usually unavailable with the time resolution of size distribution measurements.

answer: we include now the standard deviation of the RI_{eff} and the chemical composition RI in the conclusions.

reviewer comment: Page 19: Line 1: It would help to state the seasonal variability more strongly if the readers could comment whether the seasonal values or wind direction values were greater than the experimental uncertainty. As it stands, it appears somewhat subjective.

answer: The text now reads: *"In spite of the strong seasonal variability of the chemical composition at the measurement site (e.g. 86% sea-salt present in winter, 50% in summer) ..."*

reviewer comment: General comment: Please provide wavelength and RH on each of the figure and table captions- it can help the reader quickly orient themselves without having to scroll back through the text.

answer: We provide now wavelength and RH throughout.

References

Chylek, P., and J. Wong, Effect of absorbing aerosols on global radiation budget, Geophysical Research Letters, 22 (8), 929-931, 1995.

Tang, I.N., Chemical and size effects of hygroscopic aerosols on light scattering coefficients, Journal of Geophysical Research, 101 (D14), 19,245-19,250, 1996.

Response to Reviewer 2:

The authors would like to thank Anonymous Referee #2 for their helpful comments and suggestions.

According to the comments, we reconsidered and rectified our manuscript (ms). Below, we give a point to point reply. For convenience and to avoid an unnecessary inflation of this response letter, all corresponding changes in our revised ms are accordingly indicated. We refrain from listing all revised marginal fragments here. However, we present all essentially revised parts of the ms straight below our response *(“in quotation marks and in italics”).*

reviewer comment: My main concern is that, in some parts more than in others, the language needs polishing, beyond what can be expected to be caught during the ACP-language editing at the end of the publication process. I will not list all these occurrences where the English has to be approved, but give at least an already longish list in this review at “Technical comments”.

answer: A thorough language editing of the text was performed assisted by a native speaker.

reviewer comment: page 5, line 5: Information on where exactly particles entered the tubing during these experiments would be good. Just underneath the roof, close to the inlet line?

answer: For these experiments the instrument was repositioned and the inlet line where the instrument was connected before was closed and the tubing was removed from the inlet. The particles entered the tubing from somewhere middle of the measurement container. The following sentence was added to the page 5, line 4:

"The room air was measured by disconnecting the tubing from the inlet and sucking air from inside the measurement container."

reviewer comment: page 6, line 1-2: You show this polynomial only up to 400nm - although the data (blue dots) go up to 1000nm - does this mean you only used particles up to 400nm? Please add an explanation and/or prolong the line in Fig. 1.

answer: Yes, we used a polynomial fit only in the size range between 120 nm and 340 nm, as it was stated two lines before: "For the RI fit only this size range of the number size distribution was used." For clarification the text now reads:

"In the diameter range of the RI determination of 120–340 nm, the efficiency is between 0.77 and 0.67. The losses are significant here as well, but we consider this still as correctable. To have a continuous correction factor, the transmission efficiency (Fig. 1, blue dots) was fit within the diameter range of interest a polynomial line."

reviewer comment: page 6, line 24 ff: I have an idea what you did, here, but I am not entirely sure – this could certainly be formulated much clearer. What I think you did is the following: (1) - calculate TIR for a fixed RI (2) - take the value from the TIR at the diameter of the PSL particles. I guess one confusion was due to your use of the word “bin boundary diameter”. Maybe this could be defined once and then "LAS diameter" could be used instead, throughout the text, to make the text flow better? Also, this passage sounds as if there would basically only be a signal in one bin during a PSL calibration - this is most likely not the case. Describe this more clearly.

answer: The revised text now reads:

"…(TIR, the signal which the instrument measures) of the LAS for both PSL particles (TIR_{PSL}) and for particles with the desired RI (TIR_{RI}) as function of the particle diameter".

"The LAS delivers the number size distribution (n(D)) as the particle number concentration (N(D)) sorted into diameter bins: $n(D_i)= dN(D_i)/dlog(D_i)$, where i denotes the i^{th} diameter bin. These bins cover the whole measurement range of the instrument leaving no gaps. Each diameter bin has a lower and a higher boundary ($D_{i,lower}$, $D_{i,higher}$). These diameter bin boundaries correspond to the PSL calibration of the LAS. In order to recalculate the number size distribution to another RI, all bin boundary diameter has to be recalculated. This recalculation can be done by using the previously calculated TIR values: (1) For a single PSL calibration based bin diameter ($D_{i,PSL}$) the instrument response $TIR_{PSL}(D_{i,PSL})$ is looked up. (2) Now we look at the TIR values that are calculated for the desired RI. We search at which diameter ($D_{i,RI}$) we get the same instrument response as for PSL ($TIR\_RI(D_{i,RI})=TIR_{PSL}(D_{i,PSL})$) and that diameter is the recalculated bin boundary diameter. We repeat this for every diameter bin."

"The diameter recalculation is not always straightforward, because OPCs using a monochromatic laser often suffer from a non-monotonic instrument response at higher diameters (e.g., Hodkinson and Greenfield, 1965; Barnard and Harrison, 1988). This problem of non-monotonic instrument response was solved by smoothing the calculated instrumental response function by fitting a 5^th^ grade polynomial to the logarithm of both TIR_PSL and TIR_RI functions. Figure 2 shows an example how a single bin boundary diameter (D30_PSL, the 30th diameter bin border) is recalculated using another (m=1.4+0i) RI."

reviewer comment: page 8, line 15-16: Concerning possible changes in particle composition: The way you did your derivation of RI, however, was to assume that the particle chemistry was the same for all particles in one measured size distribution? Please explicitly say this here somehow, as I got confused by your remark here.

answer: We did not assume the same chemical composition for all particles in one measured size distribution for the RI derivation! In contrast, we derive an RI which matches only the real aerosol RI if all the particles have the same chemical composition in the measured size distribution (actually only the same RI, particles with different chemical composition still might have the same RI). Otherwise, if the aerosol population is described with a single RI value, it is some kind of an average value. Since we derived the RI using only the number size distribution in the 120-340nm particle size range, the derived RI corresponds to this size range as well, and has no information on the particles with diameters outside of this size range. If the RI changes significantly with the size, our derived RI might not be equal to the average RI considering the whole aerosol population. To clarify this point, the text now reads:

"The RI derived with our method is representative for the size range of 120–340 nm, which was used for the RI calculation. If we can assume that all particles in the number size distribution have the same RI, our calculated RI is the true RI. If the chemical composition of the aerosol is changing with the particle size, it is possible that the RI is also size dependent. Hence, our derived RI might differ from the average RI that corresponds for the complete aerosol population. In addition, we assumed a spherical shape of the particles and a negligible imaginary part of the RI. Therefore we term our derived RI the effective refractive index (RI_{eff}) from now on, and for later conclusions we have to keep in mind that the RI_{eff} might not be the true RI of an individual particle."

reviewer comment: page 8, line 29-30: "We used the method introduced in the sections 2.5 and 2.6 to determine the RI of this e-cigarette smoke." But in the paragraph above you said that the RI of the cigarette smoke was 1.43, based on literature (and if you would have had to determine it first you would run into issues with circular reasoning if you then would use this measurement to calibrate the LAS TIR). I assume this again is an issue with formulating the text. Please review.

answer: In this section (as noted in the titel) we wanted to verify our RI calculation method and especially the particle loss correction. This is the reason why we searched for an aerosol source, which has a known refractive index. If we now derive in the same way a RI value with our method and this RI agrees well with the literature value of the test aerosol (e-cigarette smoke in our case), then we know that our method (calculation and loss correction) works well. This is what we did here. We modified the text for clarification:

"We used the method introduced in the sections 2.5 and 2.6 to calculate the RI of this e-cigarette smoke, first with the uncorrected LAS data then with applying the above introduced (Section 2.3) LAS correction. These values can be compared to the e-cigarette smoke's literature RI value of 1.43 to check whether the LAS correction works well or not."

reviewer comment: page 9, line 4-6: Again confusing, so let me ask you again if this is what you did: When retrieving the RI for the uncorrected LAS data, you obtained an RI of 1.35, but when you corrected the measured LAS size distribution as described above and then retrieved the RI again, you got a value of 1.43, in agreement with literature. - If this is what you did, feel free to use my sentence here in the review instead of what you wrote. Your text here was hard to follow and it took me a while until I understood what you (likely) meant.

answer: Yes, this is exactly what we did. This point should be now clear after the following changes in the text:

"Without using the LAS correction on the LAS data (green lines) we get an RI of 1.35 from the best fit. This value is significantly lower than the literature RI value of 1.43 suggesting that the LAS losses had a high influence on the retrieved RI and that a correction is necessary. When we corrected the measured LAS size distribution as described above (Section 2.3), the best fit between the SMPS and the LAS data (blue lines) resulted in the RI of 1.43, which is in agreement with the literature value. This verifies our LAS correction, and we applied it on all LAS data before November 2017."

reviewer comment: page 12, first paragraph of 3.5: I would recommend to start this paragraph differently – the first sentence states something that seems not to hold once one read the list of RIs: when looking at this list and the most abundant components of the aerosol, one wonders if this really can be in good agreement, since particularly sea salt and ammonium sulphate are clearly above the value you retrieved. This all becomes much clearer further down, but I recommend to avoid confusion and to remove this first sentence or replace it with a sentence that says what you are aiming at in 3.5.

answer: The first part of the sentence was replaced:

"The aerosol chemical composition shows a strong seasonal variation at our measurement site. The dominant aerosol component is sea-salt with around 50% of the total mass in summer..."

reviewer comment: page 17, line 2: Maybe add that you expect this because scattering scales with the diameter squared

answer: We added this note:

".. as expected, because scattering increases faster than linearly as function of the particle diameter."

reviewer comment: page 18, first paragraph: You spend most of the space in this paragraph on discussing why this one value does not make sense, and the reason basically is that the underlying data is corrupted. Maybe just do not present the blue line in the figure and say up front that due to a) the strange kink in the LAS distribution and b) due to the low particle number concentration at the larger particle diameters no useful value resulted. (I'd be afraid that otherwise in the future someone might just grab that value from your figure without reading the text and use it.) Also, this lowering for particles >~350 nm, together with the bimodality you showed in Fig. 9 - could this point towards two different sources for particles? This is something you could discuss here, instead.

answer: We now removed the blue line. We modified the paragraph accordingly:

"The conspicuously lower RI_{eff} in the highest investigated size range may originate from a significantly changing chemical composition. Interestingly, sea-salt particles should dominate this higher size range, but this would result in a higher RI_{eff}. Hence, one may speculate about a coating of sea-salt particles in this special case (probably organic material with typically lower RI). The presence of a coating or a different aerosol source might also explain the bimodality of the scattering coefficient size distribution (Section 3.8)."

reviewer comment: page 6, line 26: Do you really mean an OPC (i.e., a counter) or rather an OPS (optical particle sizer)? (Check this also in the introduction, line 19 on page 2).

answer: Optical particle counters (OPCs) are not only counting but also sizing the particles. They are just for some historical reason called counters. Optical particle sizer is more recent name for the same instrument. The older name, in our opinion, is better known and therefore we would like to leave it as it is.

reviewer comment: page 8, line 9-10: This again is a strangely formulated sentence.

answer: We changed the sentence:

"The Chi function was determined for every single m value, and the minimum of this function was searched. The m value, where Chi reaches its minimum is the m we look for and we interpret as the RI of the measured aerosol."

We especially thank the reviewer for numerous technical comments! We agree with all suggested corrections and modified the text accordingly throughout.

Response to Reviewer 3:

The authors would like to thank Anonymous Referee #3 for their helpful comments and suggestions.

According to the comments, we reconsidered and rectified our manuscript (ms). Below, we give a point to point reply. For convenience and to avoid an unnecessary inflation of this response letter, all corresponding changes in our revised ms are accordingly indicated. We refrain from listing all revised marginal fragments here. However, we present all essentially revised parts of the ms straight below our response *("in quotation marks and in italics").*

reviewer comment: The authors report on aerosol refractive index observations but never mentioned that the index of refraction is a complex number. Particularly, the imaginary part of the refractive index constitutes the light-absorbing properties of the sampled aerosol. As Weller et al. (2013) reported, there is a small but significant fraction of lights-absorbing material contained in the aerosol in Antarctica. However, the authors never refer to this observation in a quantitative manner, nor they stated the assumption of a zero imaginary part of the refractive index. Furthermore, the scattering cross-section as calculated by Mie or Rayleigh-Debye-Gans theories depends on the square of the complex refractive index which includes the imaginary part. I request a discussion of the uncertainties in calculating the real part of the refractive index, when neglecting the imaginary part. The effect may be small but it should be mentioned since the imaginary part plays a crucial role in the aerosol radiation interaction.

answer: We agree about the importance of the imaginary part of the refractive index and the light absorption. However, we do not agree, that we did not mention the assumption of a zero imaginary part of the refractive index (see: Page 8, Line 7-9: "The imaginary part of the RI was kept at 0 which is an acceptable assumption considering that the absorption is very low compared to the scattering at our measurement site, average single scattering albedo at Neumayer is 0.992 (Weller et al., 2013). Page 8, Line 16-17: "The other assumption we use is that the aerosol particles are spherical and that the imaginary part of the RI is negligible." Page 13, Line 17-18: "The imaginary part of the RI was again neglected, which is surely a justified assumption, because the volume fraction of the BC never exceeded 0.1% in 2017."

Nevertheless, we agree, that a thorough discussion on the effect of the neglected imaginary part of the RI improves the manuscript. Accordingly, we modified and supplemented the text as follows:

Abstract

"Given the high average scattering albedo of 0.992 (Weller et al. 2013), we assumed that the imaginary part of the RI is zero."

Section 3.5

"The imaginary part of the RI was again neglected, which is a justified assumption, because the volume fraction of the eBC never exceeded 0.1% in 2017. This amount of eBC would add at most a ~4x10-3i imaginary value of the RI."

Section 3.7

"Finally we investigate the effect of neglecting the imaginary part of the RI on the scattering coefficient. As we have seen in Section 3.5 including the eBC in the chemical composition adds at most an imaginary part of ~$4x10^{-3}$i to the RI. We recalculated the average scattering coefficient size distribution adding this imaginary part to the RI. This gives us a highest possible estimate on the error we make if we would neglect the imaginary part of RI. It turns out that the relative difference of the scattering coefficient

size distribution considering 4x10^{-3}i RI instead of 0.0i never exceeds 1.7% irrespective of the particle diameter."

reviewer comment: Figure 3: I assume that the dashed green line refers to the LAS uncorrected best fit, please add.

answer: Yes, the reviewer is right and we corrected the figure accordingly.

reviewer comment: Figure 10: I propose to specify LAS original as LAS (m = 1.59); the term "original" suggests that data were modified, which is, however, not the case.

answer: Thanks for this reasonable suggestion that we adopt now for this and the other figures as well.

reviewer comment: When reporting on the black carbon mass concentration determined by the MAAP, the authors should use the today accepted terminology of "equivalent black carbon" (eBC); see Petzold et al. (2013).

answer: Again, we adopted the reviewers suggestion in the revised version of our manuscript and included the reference.

One year of aerosol refractive index measurement from a coastal Antarctic site

Zsófia Jurányi[1,a] **and Rolf Weller**[1]

[1] Alfred-Wegener-Institut Helmholtz Zentrum für Polar- und Meeresforschung, Bremerhaven, Germany
[a] now at: Institute for the Protection of Maritime Infrastructures, German Aerospace Center (DLR), Bremerhaven, Germany

Correspondence: Zsófia Jurányi (zsofia.juranyi@gmail.com)

Abstract. ~~Climate change model evaluations need~~

Though the environmental conditions of the Weddell Sea region and Dronning Maud Land are still relatively stable compared to the fast-changing Antarctic Peninsula, we may suspect pronounced effects of global climate change for the near future (Thompson et al., 2011). Reducing the uncertainties in climate change modeling requires inter alia a better understanding of the ~~atmospheric aerosols' optical propertiesand with this of the~~ aerosol optical properties, and for this we need accurate data on the aerosol refractive index (RI)~~of atmospheric aerosols as well~~. Due to the remoteness of Antarctica only ~~a very few data on the refractive index exists from there. In this paper we~~ very few RI data are available from this region (Hogan et al., 1979; Virkkula et al., 2006; Shepherd et al., 2018). We calculate the real refractive index of natural atmospheric aerosols from number size distribution measurements at ~~a coastal Antarctic measurement site. In our calculations we used~~ the German coastal Antarctic station Neumayer III. Given the high average scattering albedo of 0.992 (Weller et al., 2013), we assumed that the imaginary part of the RI is zero. Our method uses the overlapping size range (~~120–340~~particle diameter D between 120 and 340 nm) of a scanning mobility particle sizer (SMPS), which sizes the particles by their electrical mobility, and a laser aerosol spectrometer (LAS), which sizes the particles by their optical scattering signal at 633nm wavelength.

Based on almost a complete year of measurement~~and 2439 measurement points~~, the average effective refractive index (RI$_{\mathrm{eff}}$~~)~~, as we call our retrieved RI because of the used assumptions) for the dry aerosol particles turned out to be 1.44 ~~. This is~~ with a standard deviation of 0.08, in a good agreement with the RI value of 1.47, which we derived from the chemical composition ~~filter~~ of bulk aerosol sampling measurements. At ~~our measurement site the aerosol has a very characteristic~~ Neumayer the aerosol shows a pronounced seasonal pattern in both, number concentration and chemical composition. Despite this, ~~we could not identify any significant seasonal variability in the~~ the variability of the monthly averaged RI$_{\mathrm{eff}}$ ~~, the monthly averages remain within the range of~~ values remained between 1.40 ~~–1.50. Two~~ and 1.50. Compared to the annual mean, two austral winter months ~~June and Septemberhas a slightly higher average~~ (July and September) showed slightly but significantly increased values (1.50 and 1.47, respectively). The size dependency of the RI$_{\mathrm{eff}}$ could be determined from time averaged LAS and SMPS number size distributions measured between December 2017 and January 2018. Here we calculated RI$_{\mathrm{eff}}$ for four different particle size ranges and observed a slight decrease from 1.47 (D range 116–168 nm) to 1.37 (D range 346–478 nm).

We ~~could not identify any influence of the occurring wind direction on the retrieved~~ find no significant dependence of the derived RI$_{\mathrm{eff}}$ values on the wind direction. Thus we conclude that RI$_{\mathrm{eff}}$ ~~either. For the few examples of north winds coming from the Neumayerstation (occurs very rarely, this is the reason why the measurement site was built to the south), we don't see different values than for the other wind directions. During an artificial,~~ is largely independent on the general weather situation, roughly classified in (i) advection of marine boundary layer air masses during easterly winds caused by passing cyclones in contrast to (ii) air mass transport from continental Antarctica under southern katabatic winds. Neumayer, the only relevant contamination source, is located 1.5 km north of the air chemistry observatory, where the measurements were performed. Given that northerly winds are almost absent, the potential impact of local contamination is minimized in general. Indeed our data show no impact of local contamination on RI$_{\text{eff}}$. Just in one case a temporary high contamination episode ~~, when diesel engines were operated~~ with diesel engines operating right next to the measurement site ~~, we had an hour of constant conditions such that one RI fit was possible. This fit~~ resulted in an unusual high RI$_{\text{eff}}$ of 1.59, ~~which is most probably due to~~ probably caused by the high black carbon content of the ~~diesel engine emission. Therefore, we also assume that even during northerly wind directions we did not have significant influence from the Neumayer station~~exhaust fumes.

~~During a shorter period between 2017 December and 2018 January we used the time averaged LAS and SMPS number size distributions to get some information on the size dependency of the refractive index. The RI~~To conclude, our study revealed largely constant RI$_{\text{eff}}$ ~~was fit in 5 different particle size ranges, and we have found a slight decrease of the~~ values throughout the year without any sign of seasonality. Therefore, it seems reasonable to use a single, constant RI$_{\text{eff}}$ ~~with the particle size from 1.47 in the 116–168 to 1.37 in the 346–478 range~~value of 1.44 for modeling optical properties of natural, coastal Antarctic sub-µm aerosol.

---

1 Introduction

Atmospheric aerosols affect the radiative balance of planet Earth (e.g. Ramanathan et al., 2001): ~~directly~~ Directly by absorbing and scattering the sunlight ~~(Schwartz, 1996)~~ (e.g. Schwartz, 1996) and indirectly through modifying the microphysical properties of the clouds ~~(Lohmann and Feichter, 2005)~~(e.g. Lohmann and Feichter, 2005). The current state of the scientific knowledge on the total (direct and indirect) aerosol effect is still considered low due to the complexity of these effects (IPCC, 2014).

The refractive index (RI) of the atmospheric aerosols is a key parameter calculating their absorption and scattering and therefore essential for the global modeling of the aerosol's radiative effects. Valenzuela et al. (2018) showed that there is still clearly a need for additional and accurate measurements of the RI. There are more existing optical software packages for the optical properties of the atmospheric particulate matter and these packages extensively use RI values of the different kind of aerosols. The OPAC (Optical Properties of Aerosols and Clouds, Hess et al., 1998) package is based on laboratory measurements, whereas the HITRAN-RI (HIgh-resolution TRANsmission Refractive Indices, Massie and Hervig, 2013) package uses both laboratory and field measurements for the different included components and allows comparisons between the products using the different RIs as well. ~~Valenzuela et al. (2018) showed us that there is still clearly a need for additional and accurate measurements of the RI.~~

~~The most~~ A common method to determine the RI of aerosol particles is an indirect method: ~~the~~ The measurement of the absorption and/or scattering of the particles along with the knowledge of the particle's size. The absorption and the scattering of a single particle is determined by the particle's size, shape and RI. It is most often assumed that particles are spherical and for the theoretical calculations the Mie theory can be used.

Wex et al. (2009) determined the RI of secondary organic aerosol by selecting the particle size using a differential mobility analyser (DMA) and measuring the scattering signal using an optical particle counter (OPC). The same method was used by Hand and Kreidenweis (2002) on ambient aerosol~~, additionally~~ . Additionally they combined the measurements from an aerodynamic particle sizer as well, in order to gain information on the particles' density. Bukowiecki et al. (2011); Zhang et al. (2013); Zieger et al. (2015) used the number size distribution with parallel nephelometer and aethalometer measurements to determine the RI of ambient aerosols. A very similar method was used by Virkkula et al. (2006) ~~, where it was assumed that~~ for the Antarctic site Aboa, assuming that here the imaginary part of the RI can be neglected~~and therefore no absorption data was used~~.

Barkey et al. (2007) measured laboratory generated particles' number size distribution and ~~parallel their~~ light scattering by a polar nephelometer. They introduced an inversion algorithm to obtain the RI. A new and more exotic method is to use optical trapping combined with Mie spectroscopy to capture the RI of atmospheric aerosol samples in the 460–700 nm wavelength range by Shepherd et al. (2018). Cavity ring-down spectroscopy is a ~~commonly used~~ method to study the light extinction by aerosol particles. This method was used by Bluvshtein et al. (2012) who introduced an RI retrieval method by measuring the light extinction at two carefully selected size parameters. We have to keep in mind that all above mentioned methods are not direct measurements of the RI. All of these methods search for RI values that provide good agreement in a closure study between different measured quantities.

As we see there are plenty of existing ~~measurements on aerosol RI~~ aerosol RI measurements, but the majority of these measurements are based on laboratory generated particles and only ~~less~~ few on ambient aerosols. And if we look for RI measurements from Antarctica we can only find very few available data. Hogan et al. (1979) collected aerosol particles at the South Pole in a size range between 0.3 and 12 µm during a 4-days period and put oils with known different RIs on them until ~~the particles disappeared~~they could not see the particles in the microscope (i.e. until the applied oil's RI matched the RI of the collected particles). They have found an RI of 1.54 for these samples. Virkkula et al. (2006) derived the RI (assuming a zero imaginary part) of the ambient aerosol at coastal Antarctica during a 12-days summer campaign and got values around 1.43–1.44. Insoluble organic aerosol collected at the Clean Air Sector Laboratory of the British Antarctic Survey station Halley was analysed by Shepherd et al. (2018). ~~The samples were~~ They obtained a RI of 1.47 for samples collected on 60 consecutive days during ~~the austral summer of 2015 and they got an RI of 1.47.~~ austral summer 2015.

In this paper we would like to present continuous data on the real RI at 633 nm wavelength of the dry ambient aerosol as derived from measurements of an optical particle counter and a scanning mobility particle sizer. To our knowledge this is the first time, that such long-term RI measurements of almost one year from Antarctica is presented. With this, our study aims at better understanding of the aerosol optical properties at a place where only very few such data are available with special focus on its temporal variability. Given the distinct seasonality of the aerosol composition (see Weller et al., 2008, Figs. 4 and 5 therein), we may likewise expect a seasonality of RI. To this end, continuous year-round data of RI are necessary, in particular regarding the lack of such measurements for the Antarctic realm.

2 Method

2.1 Sampling Site

The measurements presented in this paper were performed in the Air Chemistry Observatory (SPUSO from "Spurenstoffobservatorium") of the German Antarctic station of Neumayer III ~~,~~ between February 2017 and January 2018. The SPUSO is situated at the coast of Antarctica on the Ekström ~~shelfice close to the~~ shelf-ice close to Atka Bay. This observatory is a global site of the WHO's Global Atmosphere Watch programme (World Meteorological Organisation, 2016). Detailed description of the site and of the prevailing meteorological conditions were already presented elsewhere (Wagenbach et al., 1988; Weller et al., 2008), here we only give a brief introduction to ~~the site~~it.

The SPUSO lies 1.5 km south from the Neumayer ~~station and stays~~ III station and was built on the shelf-ice which moves approximately 120 m every year to the north. The edge of the shelf-ice and with this the sea is 7-to-21 km to the north. Due to the remoteness of the measurement site, anthropogenic pollution can barely reach it ~~,~~ and the main aerosol source is the Southern Ocean. During the austral summer the sea next to the ~~shelf ice~~ shelf-ice edge and in the close Atka bay is ice free, whereas during the long ~~antarctic~~ Antarctic winter the next open water can be as far as 100 km. Towards the inside of the continent, apart from some remote nunataks there is no ice-free surface.

The only possible contamination source is the Neumayer station itself, where most of the energy is provided by diesel engines. This is the reason why the SPUSO was built 1.5 km to the south of the station in a clean air sector and its power supply is provided through a cable from the main station. At this measurement site, northerly winds are almost never present and therefore most of the time we can have a contamination free sampling. The Neumayer station is completely isolated and not ~~reachable~~ accessible during the winter season which lasts 9 months.

2.2 Experimental Setup

~~At our measurement site, the~~ The aerosol is continuously sampled through our inlet system, which has ~~it's~~ its air intake approximately 8 m above the snow surface. The inlet has an aerodynamic cut-off diameter of 7–10 µm at windspeeds of 4–10 m s^{-1} (Weller et al., 2008). Due to the heated measurement container and the low ambient temperatures, aerosol entering the measurement container is dry (relative humidity, RH<<30 %, most of the time even RH<10 %) without any additional drying. The inlet system is made of electropolished stainless steel, the individual instruments are connected to the inlet via stainless steel ~~as well~~ or/and conductive silicon tubing. The meteorological data used in this study (temperature, wind direction and speed and ambient RH) was measured directly on the roof of SPUSO.

The particle number size distribution was measured with two commercial instruments. A scanning mobility particle sizer (SMPS) consisting of an electrostatic classifier (TSI 3080) and a condensational particle counter (CPC, TSI 3776) measured in the 16–960 nm particle mobility diameter range. The SMPS was operated with 2.2 L min^{-1} sheath flow and 0.3 L min^{-1} sample flow. ~~Next to the SMPS~~ The other instrument was a laser aerosol spectrometer (LAS, TSI 3340) ~~was installed as well. This instrument~~ which detects and sizes the particles by measuring the intensity of their scattered light as they pass by the 633 nm Helium-Neon active cavity laser. The optical design and the high laser intensity ~~makes~~ enables the detection of single particles down to 90 nm diameter~~possible~~. The sample flow of the LAS was set to 0.05 L min^{-1}, the sheath flow was 0.65 L min^{-1}. The instrument measured in the size range of 90–5000 nm and was factory calibrated by Polysterene Latex (PSL) particles. Both the SMPS and LAS measured with a 10-minutes time resolution, however the LAS and the SMPS sees different particles at a time. The LAS counts all the particles which pass ~~by its~~ the laser beam whereas the SMPS performed two scans within the 10-minutes time period and is only able to detect one particle size at ~~the~~ a time, dependent on the voltage that is currently set in the instrument. Therefore if the aerosol changes significantly within 10 minutes, differences can exist between the measurements of the two instruments as well.

The particle number concentration was measured by a commercial CPC (CPC, TSI 3775) with a one-minute time resolution. A Multi-Angle Absorption Photometer (MAAP, Thermo ~~ESM Andersen~~Scientific TM Model 5012) operating at a wavelength of ~~630~~637 nm (Petzold and Schönlinner, 2004) was used to measure the ~~BC mass concentration~~ aerosol absorption during the measurement campaign. The absorption values were converted into ~~BC~~ equivalent black carbon (eBC, Petzold et al., 2013) mass concentration using a mass absorption efficiency of $6.6\,\mathrm{m^2g^{-1}}$, and were registered also once ~~in a~~ per minute. The ionic composition of the aerosol ~~is~~ was measured by a low volume Teflon/Nylon filter system, and the filters are analysed by ion chromatography. The filters were changed daily but not every day at the same time and therefore the time resolution of the ionic composition varies with ~~the~~ time. The average sampling flow was $\approx 3.5\,\mathrm{m^3h^{-1}}$, the sampled air volume varied between $30\,\mathrm{m^3}$ and $125\,\mathrm{m^3}$ in 2017. The filter sampling is automatically switched off in case of a possible contamination (snow drift, ~~wind coming from the Neumayer station, low windspeed, too high particle concentration or too high windspeed~~northerly wind direction, wind velocities below $2\,\mathrm{m\,s^{-1}}$ or above $20\,\mathrm{m\,s^{-1}}$, and exceedingly high particle number concentrations), see details in Weller et al. (2008). In this study we used the following main ionic species: NH_4^+, Na^+ NO_3^-, non sea-salt SO_4^{2-} and MSA^- (methanesulphonate). The CPC ~~, the MAAP and the filter measurements~~ and the MAAP are part of the continuous measurement program of GAW.

2.3 Correction of the LAS losses

We have collected data from both the LAS and SMPS instruments for almost one year (09.02.2017–20.01.2018). Unfortunately, during most of this time, the LAS was positioned horizontally too far away (ca. 3 m) from the inlet such ~~,~~that significant amount of particles were lost in the connecting tube. This problem was first discovered in November 2017. Right after, on the 23.11.2017, the instrument was repositioned right below the inlet in order to minimize the particle losses. ~~We used the number size distribution data in the~~ For this study, we were particularly interested in the particle diameter range between 120 and 340 nm because we used the number size distribution data in this diameter range for the RI determination (see section 2.6)~~for the RI determination, therefore~~ . Therefore, it was important to check whether or not we are able to correct for the particle losses before November 2017 in this diameter range.

Measuring the losses in the sampling line which was used before November 2017 ("old" setup) was a challenging task. ~~Our~~ At our measurement site, ~~the SPUSO, did not have any kind of particle generator~~ no particle generator was available to perform tests with, and due to the location and isolation of the station, it was also impossible to receive any equipment for the test. Our best option was to use the room air of the measurement container to quantify the ~~losses. This particle source~~ particle losses. The room air aerosol was measured by disconnecting the tubing from the inlet and sucking air from inside the measurement container. The room air provided only a low concentration~~such, that more ,~~ , so that several hours of measurement were needed. One measurement cycle included the number size distribution measurement ~~with~~ of the LAS of the room air aerosol in the "old" setup and right after removing the horizontal tube ~~another measurement~~ in the "new" setup, with the shorter, vertical tube. To make sure, that the aerosol source is stable enough during one cycle, the ~~setup was changed every~~ number size distribution measurement time was reduced to 2 times 60 s with some seconds in between to change between the setups.

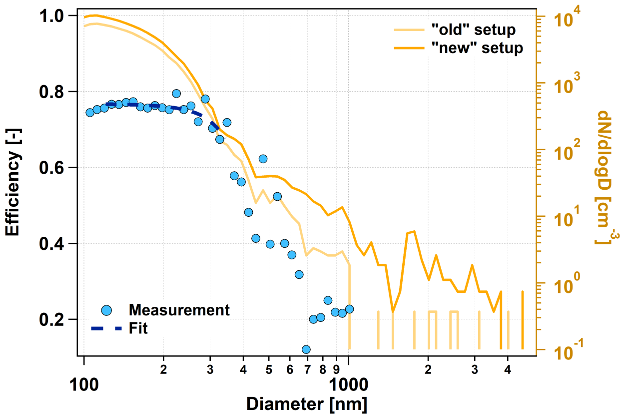

Figure 1. The quantification of the LAS losses in the sampling line. The two orange lines ~~belong~~ refer to the right axis and show the average room air number size distributions. "Old" setup: time average with the long horizontal tube, "new" setup: time average without the horizontal tube. The blue dots show the ~~penetration~~ particle transmission efficiency through the tube, the dashed dark blue line shows a polynomial fit in the diameter range which was used for the RI calculation.

All measured number size distributions were averaged separately for the "old" and the "new" setups, and the average number size distributions were compared. Figure 1 shows the results of this ~~average number size distribution~~ comparison. If one looks at ~~it~~ them (Fig. 1, orange lines, right axis) or ~~the penetration efficiency (~~at the particle transmission efficiency (the ratio between the two size distributions, Fig. 1 ~~,~~blue dots, left axis) it is obvious that the losses in the "old" sampling line are significant~~, almost~~. Almost all particles with diameters above 1 µm were lost, and therefore it is impossible to make any correction there. ~~This is the reason, why we only~~ For this reason, we have the complete number size distribution ~~until~~ up to 5 µm only after November 2017 for this study. ~~However, in~~ In the diameter range of the RI determination of 120–340 nm, the efficiency is between 0.77 and 0.67. The losses are significant here as well, but we consider this still as correctable. To have a continuous correction factor, the transmission efficiency (Fig. 1, blue dots) was fit within the diameter range of interest with a polynomial line. The blue dashed line shows this polynomial fit which was used for the correction.

2.4 Time averaging

Due to the low aerosol number concentration in Antarctica, we performed a base time averaging of one hour of all measured data. This one hour averaging still often resulted in too noisy number size distributions, such that an RI fit was impossible. The particle number concentration at our measurement site has a strong seasonal variability with much lower concentrations in winter than in summer. This ~~is why we decided to perform on top of the one hour time averaging a particle concentration dependent time averaging as well in order to keep~~ strong seasonal variability is the ~~time resolution as high as possible~~reason why in summer a much shorter time averaging period is sufficient to enable a successful RI fit. To keep the highest possible time resolution of the derived RI, we have chosen the length of the time averaging to be time dependent. And this length was determined by the actual particle concentration.

After performing many tests, we ~~concluded~~found, that the one hour averaged SMPS number size distributions, ~~that were~~ recorded during a time period with an average number concentration of at least 400 cm^{-3}~~had a good enough~~, showed an adequate signal to noise ratio for the RI calculation and no further averaging was needed. For all other cases with lower concentrations the hourly averaged data was further averaged until the number of ~~the detected particles~~ particles detected by the SMPS equaled or exceeded the particle number, which is detected during a one hour SMPS scan ~~with~~ at 400 cm^{-3} ~~concentration. With this averaging method in~~ particle concentration. In some extreme cases in winter, the measured data had to be averaged for 15 hours, whereas in summer most of the time the original one hour or ~~maybe~~ sometimes 2-hours averaging time was needed. Due to this averaging method we have the highest possible time resolution ~~but it is~~ though not constant, ~~changing in time~~but changing with time, depending on the total particle number concentration. This changing time resolution had to be taken into account for all further time average or statistical calculations.

2.5 Recalculation of the LAS number size distribution

The LAS is factory calibrated using PSL particles having an RI of 1.588 (Eidhammer et al., 2008). In order to be able to recalculate the particle number size distribution ~~to~~ for any other RI, we need to calculate the theoretical instrument response (TIR, the signal which the instrument measures) of the LAS for both PSL ~~and the desired particle RI~~ particles (TIR$_{\mathrm{PSL}}$) and for particles with the specified RI (TIR$_{\mathrm{RI}}$) as function of the particle diameter. This was done by a custom-written Mie code using the LAS wavelength of λ =633 nm and a detection angle Θ between 22 and 158 degrees with a geometry of a round detector shape.

The LAS ~~diameter bin boundaries corresponding~~ delivers the number size distribution ($n(D)$) as the particle number concentration ($N(D)$) sorted into diameter bins: $n(D_i) = \frac{dN(D_i)}{d\log(D_i)}$, where i denotes the ith diameter bin. These bins cover the whole measurement range of the instrument leaving no gaps. Each diameter bin has a lower and a higher boundary ($D_{i,\mathrm{lower}}$, $D_{i,\mathrm{higher}}$). These bin boundaries correspond to the PSL calibration ~~can be transformed to a diameter at the target RIby searching for the PSL calibration TIR value in~~ of the LAS. In order to recalculate the number size distribution to another RI, all bin boundary diameter has to be recalculated. This recalculation can be done by using the previously calculated TIR values: (1) For a single, PSL calibration based bin diameter ($D_{i,\mathrm{PSL}}$) the instrument response TIR$_{\mathrm{PSL}}$($D_{i,\mathrm{PSL}}$) is looked up. (2) Now we look at the TIR values ~~calculated with the target RIand looking up the corresponding diameter. However, this problem~~ that are calculated using the desired RI. We search at which diameter ($D_{i,\mathrm{RI}}$) we get the same instrument response as for PSL: TIR$_{\mathrm{RI}}$($D_{i,\mathrm{RI}}$) = TIR$_{\mathrm{PSL}}$($D_{i,\mathrm{PSL}}$) and that diameter is the recalculated bin boundary diameter. We repeat this for every diameter bin.

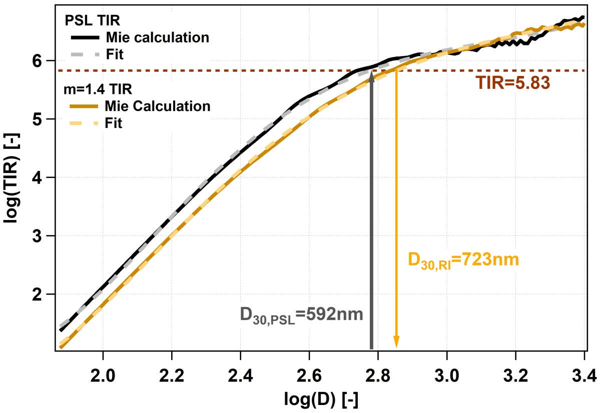

Figure 2. LAS Theoretical instrument responses for m = 1.588 + 0i (black) and 1.40+0i (orange) as function of the particle diameter. Here we show an example, how an original LAS diameter bin border ($D_{30,\mathrm{PSL}}$) was recalculated to the target RI ($D_{30,\mathrm{RI}}$).

The diameter recalculation is not always straight-forward, because OPCs using a monochromatic laser often suffer from a non-monotonic instrument response at higher diameters (e.g., Hodkinson and Greenfield, 1965; Barnard and Harrison, 1988).

~~LAS Theoretical instrument responses for m = 1.588 + 0i (black) and 1.40+0i (orange) as function of the particle diameter. Here we show an example, how an original LAS diameter bin border (D_{30}) was recalculated to the target RI (D'_{30}) is shown.~~

This problem of non-monotonic instrument response was solved by smoothing the calculated instrumental response function ~~. The smoothing was done~~ by fitting a 5th grade polynomial to the logarithm of both ~~PSL and target RI TIR~~TIR$_{\mathrm{PSL}}$ and TIR$_{\mathrm{RI}}$ functions. Figure 2 shows an example how a single bin boundary diameter (D~~$_{30}$~~$_{30,\mathrm{PSL}}$, the 30th diameter bin border) is recalculated using another RI (m = 1.4+0i)~~RI~~. The Mie calculation (solid line) and the polynomial fit (dashed line) are shown for both RIs. The 30th diameter bin border is 592 nm in our setup, using the original PSL calibration. One can read from Figure 2 that a PSL particle of this size detected by the LAS results in the same TIR as a particle with the RI of 1.4 and the size of ~~D'_{30}~~$D_{30,\mathrm{RI}}$ = 723nm. The same procedure has to be used for every bin boundary diameter and every desired index of refraction. After having the recalculated diameter borders, we can recalculate the number size distribution as well. If the original number size distribution is:

$$n_{\mathrm{PSL}}(D_{\mathrm{PSL}}) = \frac{dN(D)}{d\log(D)}\,\frac{dN(D_{\mathrm{PSL}})}{d\log(D_{\mathrm{PSL}})} \tag{1}$$

Then the recalculated number size distribution looks like this:

$$n'_{\mathrm{RI}}(D'_{\mathrm{RI}}) = \frac{dN(D')}{d\log(D')}\,\frac{dN(D_{\mathrm{RI}})}{d\log(D_{\mathrm{RI}})} = \frac{dN(D')}{\log(D'_{\mathrm{high}}) - \log(D'_{\mathrm{low}})}\,\frac{dN(D_{\mathrm{RI}})}{\log(D_{\mathrm{high,RI}}) - \log(D_{\mathrm{low,RI}})} \tag{2}$$

where ~~D'_{high}~~ $D_{\mathrm{high,RI}}$ is the upper and ~~D'_{low}~~ $D_{\mathrm{low,RI}}$ is the lower boundary of the recalculated diameter bin.

2.6 Calculation of the effective refractive index

In order to find the aerosol refractive index, the SMPS and the LAS data in the overlapping size range has to be matched. This matching was done by recalculating the LAS number size distribution using a set of different RIs and finding the one which matches the best the SMPS number size distribution at the overlapping size range. The following expression was used after Khlystov et al. (2004) to quantise the difference between the LAS and the SMPS distribution:

$$\chi(m) = \frac{1}{N} \cdot \sum_{i=N_{\mathrm{min}}}^{N_{\mathrm{max}}} \left[\log\left(n_{\mathrm{SMPS}}\left(D_i\right)\right) - \log\left(n_{\mathrm{LAS}}\left(m, D_i\right)\right)\right]^2 \tag{3}$$

The SMPS and the LAS has an overlapping size range between 90 and 950 nm, however only the range between 120 and 340 nm was used for the fit~~due to the very low particle concentration~~. The SMPS number size distribution was too noisy over 340 nm and at the lowest diameters, the LAS does not have a detection efficiency of unity. The range of the RI was chosen to be 1.3–1.8 with 0.01 steps in between. The imaginary part of the RI was kept at 0 which is an acceptable assumption considering that the absorption is very low compared to the scattering at our measurement site, with an average single scattering albedo ~~at Neumayer is~~ of 0.992 (Weller et al., 2013). The $\chi(m)$ function was determined for every single m value~~and the aerosol RI is the~~, and the minimum of this function was searched. That m ~~value where the~~ where χ ~~function~~ reaches its minimum is the m value we look for and we interpret as the RI of the measured aerosol. Those cases were omitted where the χ function did not have an explicit minimum or exceeded a limit. After manual inspection of many fit procedures this limit was set to the value of 0.02. Such cases might occur if too much noise is present in the data or if the size distribution was varying too much during the time period of one scan. Next to this numerical criterion every single scan was manually checked as well.

The RI derived with our method is representative for the ~~used overlapping size~~ particle diameter range of 120–340 nm, which was used for the RI calculation. If we can assume that all particles in the number size distribution have the same RI, our calculated RI is the true RI. If the chemical composition of the aerosol is changing with the particle size, it is ~~also~~ possible that the RI is also size dependent. ~~This we have to keep in mind for later conclusions. The other assumption we use is that the aerosol particles are spherical and that the~~. Hence, our derived RI might differ from the average RI which corresponds to the complete aerosol population. In addition we assumed a spherical shape of the particles and a negligible imaginary part of the RI~~is negligible. Due to these assumptions we call the~~. Therefore we term our derived RI the effective refractive index (RI$_{\mathrm{eff}}$) from now on, and for later conclusions we have to keep in mind that the (RI$_{\mathrm{eff}}$) might not be the true RI of an individual particle.

3 Results and discussion

3.1 Verification of the LAS correction

In order to verify the used LAS correction (see Sec. 2.3), ~~measurements~~ measurement of particles with known RI and spherical shape was necessary. The lack of any particle generator left us with not many possibilities. A commercial e-cigarette (Joytech eGo) was available at the station, and we used this to generate particles for the testing purpose. E-cigarette liquid contains glycerin, propylene glycol, water, nicotine and flavourings and the formed aerosol particles are spherical liquid droplets. Pratte et al. (2016) measured the RI of many e-cigarettes of different types and got values between 1.429 and 1.436, and therefore we assume that our generated test particles had an RI of 1.43.

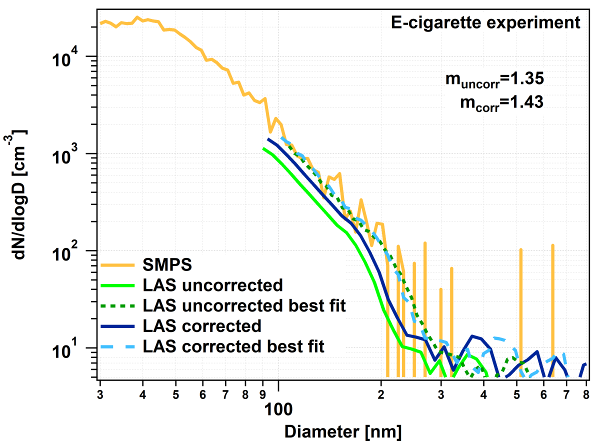

Figure 3. The E-cigarette experiment, showing the validation of our LAS correction. The orange line shows the measured SMPS number size distribution, the green lines the uncorrected LAS number size distribution (light: original, dark and dashed: best fit with m_{uncorr} calculated RI) and the blue lines (dark: original, light and dashed: best fit with m_{corr} calculated RI) are the losses corrected LAS number size distributions.

We filled a plastic bag of ≈100 L volume with particle free air, then added 2–3 puffs of the e-cigarette smoke using a small, hand-operated air pump. After that, we let the aerosol particles coagulate in the bag for 10–15~~,~~ minutes in order to let the particles reach the ~~size~~ detection diameter range of the ~~RI calculation~~LAS. The e-cigarette test was done with the same setup as the "old" measurement setup using the long vertical tube.

We used the method introduced in the sections 2.5 and 2.6 to ~~determine~~ calculate the RI of this e-cigarette smoke, first with the uncorrected LAS data then with applying the above introduced (Sec. 2.3) LAS correction. These values can be compared to the e-cigarette smoke's literature RI value of 1.43 to check whether the LAS correction works well or not. For this fit we have chosen a slightly different particle size range of 110–220 nm because the form of the number size distribution was different from the ambient one.

Figure 3 shows the results of the e-cigarette experiment. Without using the ~~above mentioned~~ LAS correction on the LAS data (green lines) we get an RI of 1.35 from the best fit. This value is significantly lower than the literature RI value of 1.43 suggesting that the LAS losses ~~have~~ had a high influence on the retrieved RI and that ~~the~~ a correction is necessary. ~~After applying the correction~~Using the losses corrected LAS size distribution, the best fit between the SMPS and the LAS data (blue lines) ~~was achieved at~~ resulted in the RI of 1.43 which ~~is in agreement~~ agrees with the literature value. This verifies our LAS correction, ~~which was applied~~ and we applied it on all LAS data before November 2017.

3.2 Sensitivity of the RI calculation on the number size distribution measurement

The accuracy of our $\mathrm{RI}_{\mathrm{eff}}$ calculation mainly depends on the measured input data's uncertainty, which is the uncertainty of the number size distribution measurements in our case. Here, we discuss the sensitivity of the derived $\mathrm{RI}_{\mathrm{eff}}$ values introduced by the measurement uncertainty. An intercomparison between many mobility particle size spectrometers showed that all of the different investigated instruments measured within an uncertainty range of ±10% (Wiedensohler et al., 2012). We use this value for our SMPS, and assume that the LAS has the same uncertainty as well.

In order to investigate the effect of this measurement uncertainty we take the worst case scenarios, by either adding 10% to the particle number concentration measured by the SMPS and subtract 10% from the LAS, or the other way around. We calculated for one month ~~of~~ measurement period the $\mathrm{RI}_{\mathrm{eff}}$ values using these modified number size distributions next to the original ones. Choosing 10% higher SMPS concentration and 10% lower LAS concentration resulted in lower calculated $\mathrm{RI}_{\mathrm{eff}}$. On average the values were 0.045 lower compared to the original values which translates into an average 3.1% error. The other scenario results in artificially high values, which turned out to be on average 0.050 and this means an error of 3.5%. This shows that even assuming the worst case scenario would cause an acceptable error, and most probably we can ~~count with~~ expect a lower uncertainty in reality.

3.3 RI calculation examples

Figure 4 shows four examples ~~about the performance~~ of the RI fitting procedure's performance in different cases. The first ~~snapshot of Figure~~example (Fig. 4a) is from the summer season when the number concentration was high enough that ~~no further averaging was necessary than the~~ a one-hour averaging period was reasonable. The orange line shows the measured SMPS scan, whereas the dark blue line shows the simultaneously measured LAS number size distribution with the factory calibration. The dark blue line lies below the SMPS line which indicates that the built-in calibration RI of 1.588 overestimates the prevailing RI. The fitting procedure verifies this and the best fit belongs to the recalculated LAS scan with the RI of 1.45 which we consider as the effective refractive index, $\mathrm{RI}_{\mathrm{eff}}$, of the dry aerosol at that time~~point~~.

Figure 4b shows a similar situation from winter with much lower particle concentrations and therefore a longer averaging time of 11 hours. The obtained RI was quite low: 1.37 in this case. An uncommon example can be seen in figure 4c when the number size distribution was trimodal. The fit was successful ~~this case as well~~again, the retrieved RI is 1.48. As the last example (Fig. 4d), we show a case where the fit was unsuccessful, we could not retrieve a valid RI. The fitting procedure returned a best fit, but the value of χ exceeded

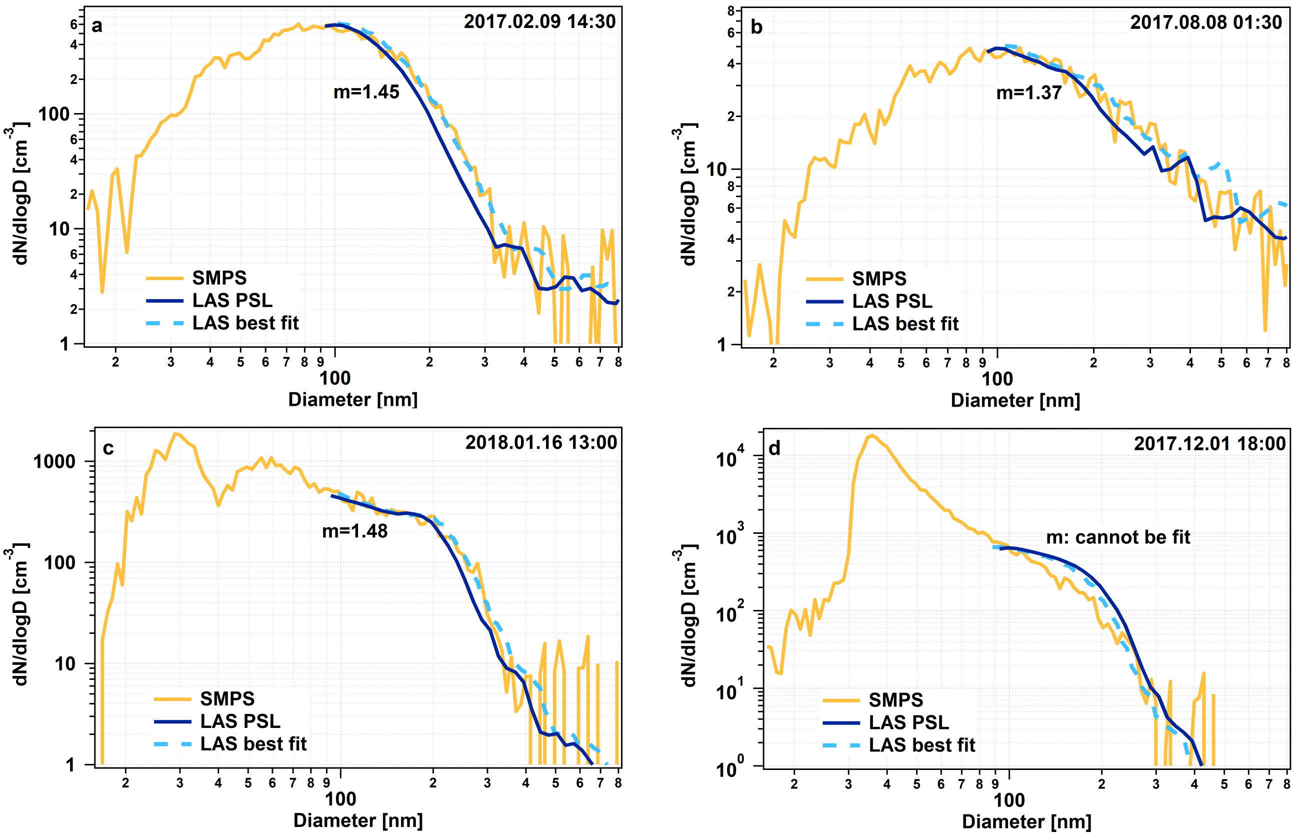

Figure 4. 4 examples on the refractive index fit performance. The orange line shows the measured SMPS number size distribution, whereas the blue lines (dark: ~~original~~PSL calibrated, light and dashed: best fit) show the LAS number size distributions.

0.02 and it is also clearly visible that this best solution does not fit very well the measured SMPS number size distribution. The reason why the fit did not work in this case was that the aerosol population was significantly changing within the duration of the SMPS scan. During the first half of the scan an aerosol plume with very high concentration reached the instruments. This appears in the SMPS scan as a very high fraction of small particles, because the SMPS selected and measured the smaller particles during the first half of the scan. Contrary, the LAS captures all particles with different diameters at the same time, and therefore this event appears as an elevated overall concentration. This was an extreme and exceptional situation where some ~~construction~~ unavoidable construction work was done around the SPUSO using machines powered by diesel engines.

3.4 Seasonal variability and mean value of the refractive index

We have collected data during almost a complete year (from 09.02.2017 to 20.01.2018), ~~and this gave~~ giving us the unique possibility to calculate the long-term RI$_{\mathrm{eff}}$ and ~~be able~~ to analyze its seasonal variability. Figure 5 shows this seasonal variability, where ~~the statistics of the montly~~ some statistical values of the monthly RI$_{\mathrm{eff}}$ ~~is~~ are presented. The gray circles show the monthly mean values with the standard deviation (Stdev) as error bars, the black sticks the medians and the gray sticks the 25$^{\mathrm{th}}$ and 75$^{\mathrm{th}}$ percentiles. The orange bar chart belongs to the right axis and indicates the number of the RI$_{\mathrm{eff}}$ values that could be retrieved for the ~~according~~ corresponding month. The same data is also shown in Table 1 ~~, and it is~~ complemented with the yearly mean values~~as well~~.

The ~~average~~ mean RI$_{\mathrm{eff}}$ during our complete measurement period was ~~1.438 and the median is also close with the value of 1.414~~1.44 with a comparable median of 1.41. As already mentioned, there are ~~are only a~~ only very few other RI measurements from Antarctica. Virkkula et al. (2006) calculated the RI values from number size distribution and scattering coefficient measurements at the Finnish Antarctic summer station ~~of~~ Aboa. Aboa is situated approximately 300 km to the west of the Neumayer station and lies a little further away from the sea. These measurements were performed in the summer of 2000 during a ~~12-days~~ 12-day period. They ~~got an average~~ found a mean RI of 1.454 at $\lambda = 550$ nm and 1.460 at $\lambda = 700$nm wavelength excluding a nucleation event ~~during they got unrealistic low~~ where unrealistically

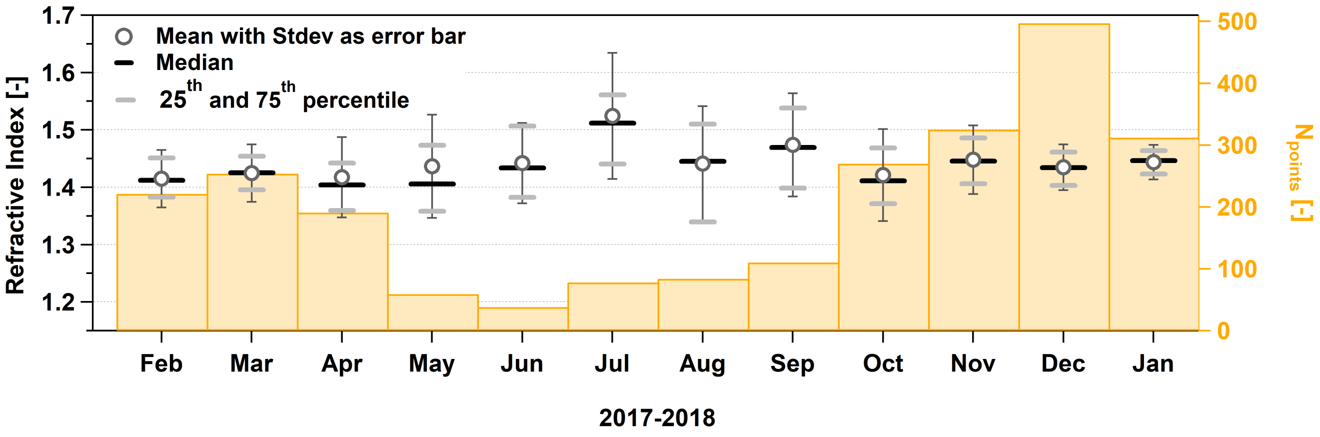

Figure 5. The monthly averages (with error bars as the standard deviation), medians and percentiles of ~~the effective~~ RI$_{\mathrm{eff}}$ from the coastal Antarctica, measured at λ=633 nm for dry aerosol particles. The orange bars refer to the right axis and show the number of ~~successfull~~ successful RI retrievals in the corresponding month.

Table 1. The monthly and yearly ($\sum$) averages, standard deviations (Stdev), medians and percentiles of the RI$_{\mathrm{eff}}$ from coastal Antarctica, measured at λ=633 nm for dry aerosol particles.

Month	25th percentile	Median	75th percentile	Mean	Stdev	N_{points}
Feb	~~1.382~~1.38	~~1.412~~1.41	~~1.451~~1.45	~~1.412~~1.41	0.05	221
Mar	~~1.395~~1.40	~~1.425~~1.43	~~1.454~~1.45	~~1.422~~1.42	0.05	254
Apr	~~1.359~~1.36	~~1.404~~1.40	~~1.442~~1.44	~~1.410~~1.41	0.07	191
May	~~1.358~~1.36	~~1.401~~1.40	~~1.473~~1.47	~~1.423~~1.42	0.09	59
Jun	~~1.382~~1.38	~~1.434~~1.43	~~1.507~~1.51	~~1.439~~1.44	0.07	38
Jul	~~1.440~~1.44	~~1.512~~1.51	~~1.561~~1.56	~~1.501~~1.50	0.11	78
Aug	~~1.339~~1.34	~~1.445~~1.45	~~1.510~~1.51	~~1.437~~1.44	0.10	84
Sep	~~1.398~~1.40	~~1.469~~1.47	~~1.538~~1.54	~~1.470~~1.47	0.09	110
Oct	~~1.371~~1.37	~~1.411~~1.41	~~1.468~~1.47	~~1.424~~1.42	0.08	270
Nov	~~1.406~~1.41	~~1.446~~1.45	~~1.486~~1.49	~~1.449~~1.45	0.06	325
Dec	~~1.403~~1.40	~~1.434~~1.43	~~1.461~~1.46	~~1.435~~1.44	0.04	497
Jan	~~1.423~~1.42	~~1.446~~1.45	~~1.464~~1.46	~~1.442~~1.44	0.03	312
$\sum$	~~1.369~~1.37	~~1.414~~1.41	~~1.462~~1.46	~~1.438~~1.44	0.08	2439

low values (lower than the RI ~~value~~ of water) ~~RI values~~were derived. Our average RI values have a very good agreement with their average RI values, and this agreement is even better ~~if we only compare our average~~ considering only our mean RI$_{\mathrm{eff}}$ value from January (~~1.446)with theirs~~1.45).

~~If we look at the montly~~ Concerning the monthly averages, it is interesting~~to see~~, that in spite of the existing strong seasonal variability of both the aerosol concentration (Jaenicke et al., 1992; Weller et al., 2011) and chemical composition (Wagenbach et al., 1988) the RI does not or only slightly show ~~the same behaviour: the~~ a comparable behaviour: The monthly averages of RI$_{\mathrm{eff}}$ ~~stay~~ remain quite constant and remain within the range of 1.40–1.50. There are two winter months with higher RIs: July with a mean of ~~1.501~~ 1.50 and September with ~~1.470~~1.47. These values are significantly different from the yearly mean (determined by using a statistical T-test with a significance level of 0.01). In both ~~of these months~~ cases we have only relatively few data-points due to ~~the extremely low concentrations and therefor~~ extremely low particle concentrations and therefore we can only speculate on the reason for the slightly higher values. In winter the fraction of sea salt is higher than in summer and sea salt has a ~~little bit~~ slightly higher RI than the other salts present in the aerosol phase (see ~~more discussion in~~ Sec. 3.5).

The monthly RI$_{\mathrm{eff}}$ distributions are quite narrow~~, there is not a big scatter in the data~~. However, due to the needed long ~~time averaging~~ averaging time between 1 and 20 hours~~we might miss the possibly existing~~, a potential higher short-term variability may not be represented. Although the Stdev of RI$_{\mathrm{eff}}$ comprising the whole measurement period is

0.08, we observed a statistically significant seasonality in the monthly data. The winter months (June to September) seem to have a higher scatter (Fig. 5 gray sticks) ~~, but this might be due to the higher~~ and higher Stdev values (0.11 in July vs. 0.03 in January, Fig. 5 error bars). We found a similar tendency in the chemical composition with higher variability during the austral winter compared to summer. This might be one reason for the higher scatter in the RI_{eff} values, apart from probably higher uncertainty of the fitting method ~~because of the extreme low concentrations and longer averaging time~~due to extremely low wintertime particle number concentrations.

3.5 Link to the chemical composition

The ~~average RI_{eff} of 1.438 is in a good agreement with what we would expect, if we look at the chemical composition of the aerosol at the SPUSO~~aerosol chemical composition shows a strong seasonal variation at our measurement site. The dominant aerosol component is sea-salt ~~at this site,~~ with around 50 % of the total mass in summer and 86 % in winter (Weller et al., 2008). ~~Next to it~~While negligible during winter, biogenic sulphur ~~is also a very important component when it~~ aerosol reaches its annual maximum in ~~summer~~ austral summer between January and March (Minikin et al., 1998). At our investigated wavelength of 633 nm, sea-salt has an RI of 1.49 (Shettle and Fenn, 1979), sulfuric acid 1.42 (Palmer and Williams, 1975), ammonium sulphate 1.53 (Toon et al., 1976), ammonium bisulphate 1.47 (Chylek and Wong, 1995), sodium nitrate 1.46 (Cotterell et al., 2017), ammonium nitrate 1.52 (Toon et al., 1976), MSA 1.43 (Virkkula et al., 2006) and black carbon 1.75+0.43i (Hess et al., 1998).

The chemical composition was determined from the daily filter measurements of the ionic composition and from the ~~BC~~ eBC measurement of the MAAP. The mass concentration of the dominant component of sea salt was calculated from the Na^+ ion. It was assumed that NH_4^+ ~~forms ammonium nitrate andthe rest of , which cannot be neutralised, can be found as sodium nitrate. In the rare case when there is enough to completely neutralise the , the rest of the~~ is preferentially present as ammonium sulphate ($(NH_4)_2SO_4$) and/or ammonium bisulphate (NH_4HSO_4) salt due to the high nss-SO_4^{2-}/NH_4^+ ~~forms ammonium sulphate~~ratio of around 11.2±8 (annual mean ± Stdev). In addition, formation of ammonium nitrate (NH_4NO_3) has to be considered. Part of the nitrate can also be bound as $NaNO_3$. The remaining SO_4^{2-} was assumed to be ~~found~~ present as sulfuric acid.

We do not have any information on the organic carbon mass fraction for our measurement period, and therefore we could not include this component into the calculation. However, previous water soluble organic carbon (WSOC) mass concentration measurements (Weller et al., 2015) showed, that in the austral summer of 2011 the WSOC average mass fraction was less than 3% and therefore we believe that organic carbon does not have a significant influence on the resulting RI. Using this chemical composition and assuming that the aerosol is homogeneously and internally mixed, the RI can be calculated from the volume fraction and the RI of the individual components. The imaginary part of the RI was again neglected, which is ~~surely~~ a justified assumption, because the volume fraction of the ~~BC~~ eBC never exceeded 0.1 % in 2017. This amount of eBC would add at most a $\approx 4 \cdot 10^{-3}$i imaginary value to the RI.

The average RI calculated from the chemical composition in 2017 becomes 1.47, and ~~as it was already mentioned,~~ is in a good agreement with the optically retrieved RI_{eff} of ~~1.438. The slight difference may come next to~~ 1.44. The reason for the slight discrepancy might be caused by the used assumptions~~from the fact that we used~~ . In addition and in contrast to the bulk chemical composition~~to estimate the RI whereas we use only the~~ , the RI calculation derived from the SMPS and OPC data are based on a limited size range between 120 and 340 nm~~for the fit and, as we will see later for a longer averaged time period (~~. As discussed later in Section 3.7~~) that the~~ , RI changes slightly with the particle size.

~~If we average the chemical composition derived~~ Finally, we calculated RI separately for ~~the~~ summer (November to February) and ~~for the~~ winter (March to October) ~~we get slightly higher values for the austral winter with 1.482 than for the austral summerwith 1.454. This is due the higher sea-salt fraction in winter , which has a higher~~ from the aerosol chemical composition. We found higher RI values of 1.48 during austral winter compared to 1.45 during summer. This may be caused by the much higher sea salt aerosol portion during winter with the highest RI among the ~~components. This might explain as well why we have gained with the fitting procedure a slightly~~ ionic compounds. Note also the significantly higher RI_{eff} values for ~~two winter months~~ the winter months July and September (Fig. 5).

3.6 ~~Influence~~ Impact of ~~the~~ general weather situation and local contamination

~~As we have already mentioned before the main station of Neumayer is a possible contamination source and we would like to investigate if this contamination source effects our derived RI_{eff} values or not. Therefore, first we check~~ Neumayer station is situated 1.5 km north of the measurement site, thus contamination during northerly winds, but also when the wind speeds are very low, has to be considered. We start with examining whether the actual wind direction influences our data ~~and then we show~~ in general, followed by a case study when diesel engines were operated right next to the measurement site. Contamination is mainly associated with high concentrations of black carbon. Black carbon has an RI of 1.75+0.43i (Hess et al., 1998) which is considerably higher than of any other natural chemical components of the aerosol. Note also the distinct imaginary part of the RI.

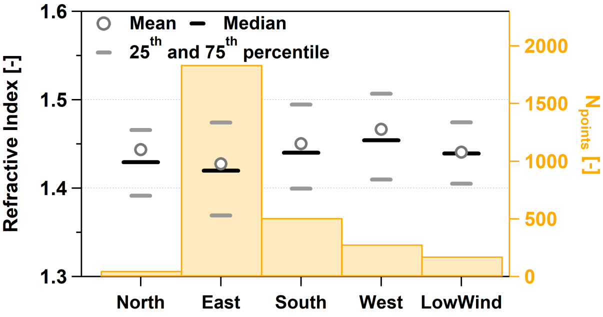

Figure 6. The averages, medians and percentiles of the RI_{eff} from the coastal Antarctica separated by the wind direction, measured at λ=633 nm for dry aerosol particles. The orange bars refer to the right axis and show the number of successful RI retrievals.

The prevailing wind direction at the SPUSO is east, ~~often~~ associated with high wind speeds ~~over~~ above $10\,\mathrm{ms^{-1}}$, ~~and wind speeds over~~ frequently exceeding even $20\,\mathrm{ms^{-1}}$~~are restricted to this direction~~. Easterly wind directions, especially if they are accompanied with high wind speeds, are characteristic for the impact of passing cyclones and marine air entry. The second frequent wind direction is south, ~~but here the wind speeds are much lower, always stay~~ with wind speeds generally below $10\,\mathrm{ms^{-1}}$~~caused by katabatic flows~~. This weather situation is characteristic for advection of more continental air masses by katabatic winds. Westerly winds ~~can also be present, even~~ are usually caused by low-pressure systems in the southern Weddell region and associated with moderate winds speeds between 10 and $20\,\mathrm{ms^{-1}}$~~, but northerly winds are almost never presentat the measurement site(König-Langlo et al., 1998)~~. Northerly winds are virtually absent (König-Langlo et al., 1998) and if present, mark a period of potential contamination from the station. We have separated the RI_{eff} data according to ~~the~~ different wind direction sectors ~~, in order to see if the different air-masses carry particles with different RI or not. We~~ to examine whether different air masses are associated with particles showing different RI. To this end, we defined the wind direction sector between 315° and 45° as north, 45° and 135° as east, 135° and 225° as south and 225° and 315° as west. ~~All the data~~ We categorized all data associated with wind speeds below $2\,\mathrm{ms^{-1}}$ ~~was categorized separately as a condition with low or no wind, and has the possibility that contamination reached the measurement site as well. Our measurement period was not exceptional: during most of the time when RI_{eff} could be retrieved, easterly winds were present (orange bars~~ separately (LowWind in Figure 6). ~~We got many points during south and west wind, conditions with low wind did not occur often and only a very few points could be fit during winds coming from the northern sector, but we still have enough data points (49) to analyse them further.~~

Overall, our measurement period was representative and meaningful for each individual sector, even for the inherently few data related to northerly wind directions. Figure 6 shows the ~~the~~ RI_{eff} values, sorted according to the ~~wind direction~~mentioned categories. The gray circles show the time averages, the black sticks the medians, the gray sticks the 25th and ~~75~~5th percentiles. ~~We do not see any~~ In summary, no significant dependency of RI_{eff} on the wind direction ~~, RI_{eff} seems to be stable and independent on from which direction the wind blew.~~

~~The main contamination source, the Neumamyer station is situated 1.5 north of the measurement site, therefore there is a possibility of contamination from this sector or for the cases when the wind speeds are very low. The contamination could origin from the diesel engines used for production of heat and electricity. This would be most probably associated with high concentrations of black carbon. Black carbon has an RI of 1.75+0.43i (Hess et al., 1998) which is much higher than of any other chemical components of the aerosol and has also a significant imaginary part. Therefore it is probable that in case of a present contamination the retrieved~~ or wind speed is observable. We conclude that the general weather situation, just as local contamination, has no impact on RI_{eff} ~~gets higher as well.~~

~~Figure 6 does not show any elevated values for the cases when the wind is low or coming from north. Therefore we assume that we do not have any significant contamination even in these cases or the contamination~~. Even adverse wind condition associated with potential contamination from the exhaust fumes of the main station did not cause any significant change of RI_{eff}. ~~It is also possible, that exactly those situations could not fit successfully when contamination was present, however we did not even see an increased unsuccessful number of fits during winds from north or low wind speed values.~~

In order to further investigate the problem of the contamination we performed a case study ~~from a~~ on a time period when planned contamination reached the SPUSO. This was the same construction event which was already shown in Fig. 4d as an example for an unsuccessful fit when the aerosol was changing too fast. On the day of 01.12.2017, diesel engine powered machines were in operation in the very close vicinity of the measurement site.

Figure 7 shows the particle number concentration (green) and the black carbon mass concentration (black) as measured by the CPC and MAAP, respectively during this construction episode. The highest concentrations were present during the morning and the late afternoon even exceeding ~~6×10^6~~$6\cdot10^6\,\mathrm{cm^{-3}}$ and $8\,\mu\mathrm{g\,m^{-3}}$ which are 3–4 orders of magnitude higher than the values without contamination (Weller et al., 2011, 2013). Unfortunately, these concentrations changed very fast, depending on whether the engine emissions were directly reaching our inlet, and therefore

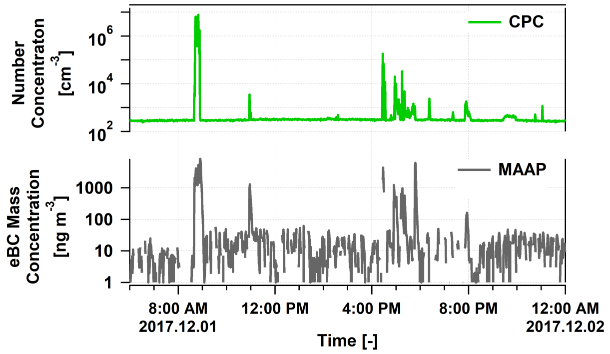

Figure 7. The particle number concentration (green) and the equivalent black carbon mass concentration (black) measured on 01.12.2017

most of the time, we were not able to perform a fit for the RI. We have only one single scan when the concentration was stable enough and elevated, ~~that we can~~ allowing us to assume that we determined $\mathrm{RI_{eff}}$ for a contaminated situation.

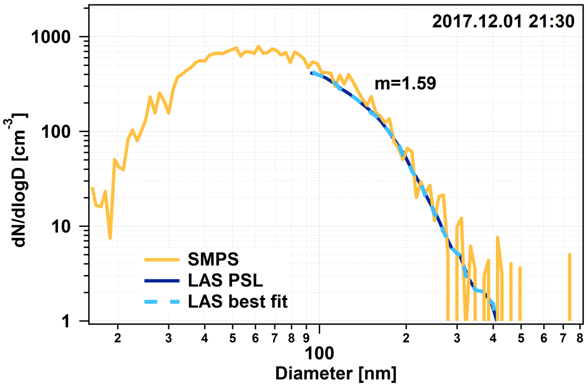

Figure 8. A successful RI fit from 01.12.2017 with high contamination present

Figure 8 shows this fit with the retrieved RI of 1.59. One can see that the original LAS scan fits already very well, which means that the RI of the factory calibration of PSLs give us a good solution. This retrieved $\mathrm{RI_{eff}}$ is significantly higher than the values we normally got. We can assume that the increased black carbon concentration caused this effect, and increased RI values might be an indicator for strong contamination at this site. This time period, and any other time period with known contamination was removed from the statistical calculations.

3.7 Size dependent contribution to the scattering

~~Having the data of the complete number size distribution (from 16 to 5000) and the $\mathrm{RI_{eff}}$, gives us the possibility to calculate the different sized particles' contribution~~ In the following we will calculate the contribution of the particles with different sizes to the scattering coefficient. Unfortunately, the LAS data was not usable above 600 nm during the time period when the particle losses were ~~significant~~ high, and therefore we can only do these calculations for an almost ~~2 months~~ 2-months long summer period (01.12.2017-20.01.2018) when the LAS was installed right below the aerosol inlet. It was assumed that the derived $\mathrm{RI_{eff}}$ is ~~usable~~ valid along the complete number size distribution (~~see section~~between 16 ~~3.8 where we speculate on the size dependence of the RI~~nm and 5000 nm) and that the particles are spherical and ~~the~~ thus Mie calculation can be used for the determination of the single particle scattering at the wavelength of 633 nm. The scattering coefficient size distribution of the dry aerosol was calculated as follows:

$$\frac{d\sigma_s(D)}{d\log D} = C_s\left(D, \lambda, m\right) \cdot \frac{dN(D)}{d\log D} \qquad (4)$$

where σ_s is the scattering coefficient in m^{-1}, m is the derived, time dependent $\mathrm{RI_{eff}}$ without a unit and C_s is the scattering cross section of the ~~particles~~individual particles in m^2. To calculate C_s we used our custom written Mie code.

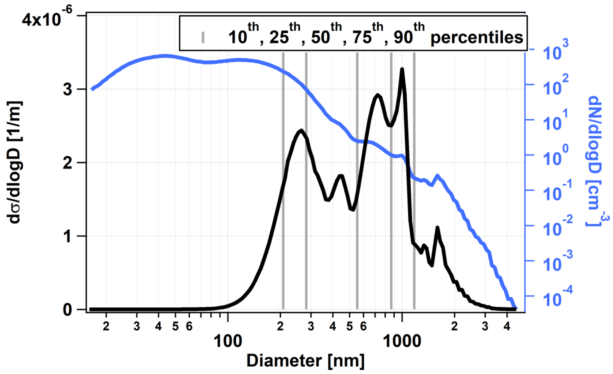

Figure 9. The average dry scattering coefficient size distribution (black line) at 633 nm wavelength and the corresponding ~~cumulative~~ particle number size distribution (~~dark green~~ blue line, right axis) as function of the particle diameter. The ~~light green~~ grey lines show the 10th, 25th, 50th, 75th and 90th of the scattering coefficent distribution.

Figure 9 shows the time average of $d\sigma_s(D)/d\log D$ as function of the particle diameter. ~~The cumulative distribution (dark green~~ Next to it, the average number size distribution (blue line, right ~~axis)~~ logarithmic axis) for the same time period is also shown. As we can see, particles smaller than 100 nm or larger than 3 µm do not contribute significantly to the scattering. 80% of the total scattering amount come from the size range between 208 and 1170 nm. Interestingly, the distribution is multimodal, having two main peaks around 260 and 860 nm. The median of the distribution is at 550 nm which is much higher than the median of the number size distribution (64 nm), as expected, because scattering increases faster than linearly as function of the particle diameter. The average number size distribution is also multimodal with two distinct peaks around 40 nm and 140 nm. Considering the time evolution and not temporal averages we see, that these two peaks, as well as the two main peaks of the scattering coefficient size distribution, are often present simultaneously. In conclusion, the bimodality is not the product of time averaging of single modes appearing at different times.

Finally we investigate the effect of neglecting the imaginary part of the RI on the scattering coefficient. As we have seen in Section 3.5 including the eBC in the chemical composition adds at most an imaginary part of $\approx 4\cdot10^{-3}$i to the RI. We recalculated the average scattering coefficient size distribution adding this imaginary part to the RI. This gives us a highest possible estimate on the error we make if we would neglect the imaginary part of RI. It turns out that the relative difference of the scattering coefficient size distribution considering $4\cdot10^{-3}i$ RI instead of 0.0i never exceeds 1.7 % irrespective of the particle diameter.

3.8 Size dependence of the refractive index

~~We have a possibility to investigate the particle size dependence of the RI. We can only use again~~ To examine the dependence of $\mathrm{RI}_{\mathrm{eff}}$ on the given particle size distribution, we again have to restrict the time period to (01.12.2017-20.01.2018) when the LAS~~was already repositioned and the~~ 's particle losses were minimised. During this period we have an SMPS–LAS overlapping size range between 120 and 900 nm. If we calculate the ~~time average for~~ temporal average over this complete time period, most of the noise ~~will be~~ is averaged out as well, ~~such~~ so that we can use ~~this complete~~ most of this overlapping size range for the RI fit. ~~This size range consists then of high enough measurements points such that it can~~ Moreover, the overall size distribution range can now be divided into ~~more subranges and we can perform the RI fit separately in these. The resulted RIvalues will describe the particles with the particle sizes of the corresponding size range~~4 subranges suitable for separate $\mathrm{RI}_{\mathrm{eff}}$ calculations, representative for the corresponding subrange (Fig. 10).

Figure 10 shows the time averaged LAS (gray line) and SMPS (black line) number size distributions. We have chosen the following particle size ranges for the separate RI fit~~and with this for the size dependency investigation~~: 117–168 nm, 168–241 nm, 241–346 nm ~~,~~ and 376–478 ~~and 478–710~~ nm ~~. We have chosen these ranges such~~ensuring, that we have a similar number of size distribution measurement points for the fit procedure in ~~every range. The coloured lines in Figure 10 show the best~~ ~~$\mathrm{RI}_{\mathrm{eff}}$~~ ~~fits~~each of the size ranges.

With the increasing particle size, we needed to apply a lower RI in order to have the best match between the LAS and the SMPS. In the first range we got an $\mathrm{RI}_{\mathrm{eff}}$ of 1.47, in the second 1.45, in the third 1.43 ~~,~~ and in the fourth 1.37~~and in the fifth 1.30~~. According to ~~this result the RIhas a particle size dependence, and it is decreasing with the diameter in the investigated size range. The decrease in~~ Figure 10 the $\mathrm{RI}_{\mathrm{eff}}$ decreases slightly within the first 3 ~~size ranges is a slight decrease and getting steeper in the last two ranges.~~ subranges of particle diameter ($\mathrm{RI}_{\mathrm{eff}}$ between 1.47 and 1.43), but more pronounced for the highest range ($\mathrm{RI}_{\mathrm{eff}} = 1.37$)

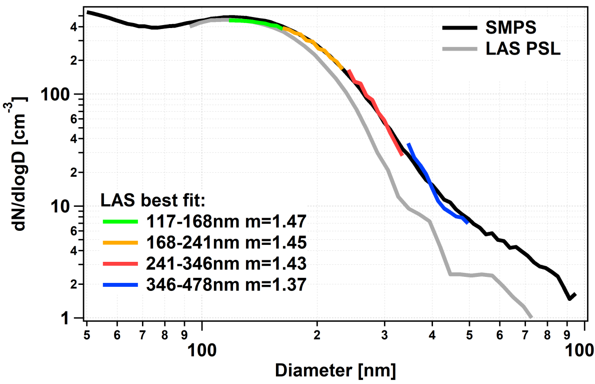

Figure 10. The average dry aerosol number size distribution measurements during December 2017 and January 2018 as measured by the SMPS (black line) and the LAS (gray line). The coloured lines show the ~~5~~ 4 individual RI ~~fitting procedures~~ fits using ~~5~~ 4 different particle size ranges.

The ~~RI in the last diameter range of 478–710 became 1.30 which is surrealistically low, even lower than 1.33, the RI of water. We have to question the quality of the fit here, even looking at the fit with the naked eyes we see that even at the best choice of RI the LAS and the SMPS do not match well. The~~ ~~χ~~ ~~value with 0.0018 is also close to the limit of 0.02 which was the limit we have set for the acceptance of a fit. One reason can be, that in this diameter range, the theoretical instrument response function is less steeper than for the lower diameters (See Figure 2) and therefore a small error in the measured scattering or in the polynomial fit of the TIR can already cause greater error in the diameter recalculation. Another possible explanation for the unrealistically low RIis the unusual shape of original LAS number size distribution in the highest chosen size range. The LAS number size distribution has a strange dip around 450~~conspicuously lower $\mathrm{RI}_{\mathrm{eff}}$ in the highest investigated size range may originate from a significantly changing chemical composition. Interestingly, sea-salt particles should dominate this higher size range, but this would result in a higher $\mathrm{RI}_{\mathrm{eff}}$. Hence one may speculate about a coating of sea-salt particles in this special case (probably organic material with typically lower RI). The presence of a coating or a different aerosol source might also explain the bimodality of the scattering coefficient size distribution (Section ~~particle diameter. This dip is often present on the single scans as well, and we cannot think of any reasons why the size distribution should have such a shape and hence we think that it might be~~

~~a systematical measurement error of the LAS. Therefore we consider the 1.30 RI value in the highest size range invalid~~3.7).

4 Conclusions

We have calculated the ~~aerosol real RI~~ real RI for dry natural aerosol at a coastal Antarctic measurement site using the overlapping size range of two ~~instrument~~ instruments measuring the number size distribution in two different ways: optically and ~~selecting the particles by their~~ by electrical mobility. The yearly average (±Stdev) of the RI was calculated based on the data from almost a complete year and turned out to be 1.44 ~~.~~ (±0.08). This average is in very good agreement with the RI value of 1.47 which we derived from filter based chemical composition measurements. The good agreement shows that at least for coastal Antarctica this method reliably delivers the RI values without the additional effort of a chemical characterization of the aerosol.

Based on this, we ~~suggest~~ recommend this single, ~~time~~ temporally constant refractive index value for ~~optical modeling . It would be interesting to determine the geographical borders of this value's validity~~modeling of aerosol optical properties. In this context we suggest supporting investigations to examine the validity of this approach and the usage of seasonal independent RI_{eff} values for the Antarctic region.

In spite of the strong seasonal variability of the ~~aerosol~~ chemical composition at the measurement site (e.g. 86% sea-salt present in winter, 50% in summer), we could not identify a ~~proper~~ corresponding seasonal trend of the RI, which is in good agreement with ~~what we assume from the filter based chemical composition~~ RI derived from the chemical composition of the present aerosol. We conclude that the given high variability of the ionic composition of the aerosol typical for coastal Antarctica causes only minor variability in associated RI values. We could not find any significant influence from the wind direction either. ~~Even those very few RI values, when the calculation was possible and the air was coming from the contaminated sector of the Neumayer station, were not different from the cases with other wind directions. We conclude therefore, that our data-set is not significantly influenced by contamination~~We conclude that the general weather situation, just as local contamination, has no significant impact on RI_{eff}.

~~Unfortunately, most of the time the LAS was positioned too far away from the inlet and therefore suffered from too high particle losses. These losses could not be corrected for particle sizes larger than 600~~In forthcoming related investigations at Neumayer, a year-round optical closure experiment is planned. For this, the size range between 16 nm and ~~therefore the complete number size distribution until~~ 5 µm ~~could be analysed and discussed only for a shorter summer period. During this period we could not measure the scattering coefficient because of our Nephelometer's failure. Therefore we could not perform an optical closure study between the number size distribution and the scattering coefficient using the calculated RIs. Future collected data will make this study possible~~as well aerosol scattering coefficients by integrated nephelometer measurements will be employed.

Data availability. Data reported here are available at https://doi.pangaea.de/10.1594/PANGAEA.899429 and https://doi.pangaea.de/10.1594/PANGAEA.899430 for scientific purposes.

Author contributions. ZJ has performed the measurements, analysed and interpreted the data and wrote the manuscript. RW built up the measurement site, supervised the measurements and the data analysis and reviewed and edited the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. The authors would like to thank all members of the 37th overwintering team of the Neumayer III station for their support and for being a great group. The first author of the paper would like to express her gratitude to her brother for his support during the harsh winter months in Antarctica.

References

Barkey, B., Paulson, S. E., and Chung, A.: Genetic algorithm inversion of dual polarization polar nephelometer data to determine aerosol refractive index, Aerosol Science and Technology, 41, 751–760, https://doi.org/10.1080/02786820701432640, 2007.

Barnard, J. C. and Harrison, L. C.: Monotonic responses from monochromatic optical particle counters, Applied Optics, 27, 584–592, https://doi.org/10.1364/AO.27.000584, 1988.

Bluvshtein, N., Flores, J. M., Riziq, A. A., and Rudich, Y.: An approach for faster retrieval of aerosols' complex refractive index using cavity ring-down spectroscopy, Aerosol Science and Technology, 46, 1140–1150, https://doi.org/10.1080/02786826.2012.700141, 2012.

Bukowiecki, N., Zieger, P., Weingartner, E., Jurányi, Z., Gysel, M., Neininger, B., Schneider, B., Hueglin, C., Ulrich, A., Wichser, A., Henne, S., Brunner, D., Kaegi, R., Schwikowski, M., Tobler, L., Wienhold, F. G., Engel, I., Buchmann, B., Peter, T., and Baltensperger, U.: Ground-based and airborne in-situ measurements of the Eyjafjallajökull volcanic aerosol plume in Switzerland in spring 2010, Atmospheric Chemistry and Physics, 11, 10 011–10 030, https://doi.org/10.5194/acp-11-10011-2011, 2011.

Chylek, P. and Wong, J.: Effect of absorbing aerosols on global radiation budget, Geophysical Research Letters, 22, 929–931, https://doi.org/10.1029/95GL00800, 1995.

Cotterell, M. I., Willoughby, R. E., Bzdek, B. R., Orr-Ewing, A. J., and Reid, J. P.: A complete parameterisation of the relative humidity and wavelength dependence of the refractive index of hygroscopic inorganic aerosol particles, Atmospheric Chemistry and Physics, 17, 9837–9851, https://doi.org/10.5194/acp-17-9837-2017, 2017.

Eidhammer, T., Montague, D. C., and Deshler, T.: Determination of index of refraction and size of supermicrometer particles from light scattering measurements at two angles, Journal of Geophysical Research: Atmospheres, 113, https://doi.org/10.1029/2007JD009607, 2008.

Hand, J. L. and Kreidenweis, S. M.: A new method for retrieving particle refractive index and effective density from aerosol size distribution data, Aerosol Science and Technology, 36, 1012–1026, https://doi.org/10.1080/02786820290092276, 2002.

Hess, M., Koepke, P., and Schult, I.: Optical properties of aerosols and clouds: the software package OPAC, Bulletin of the American Meteorological Society, 79, 831–844, https://doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2, 1998.

Hodkinson, J. R. and Greenfield, J. R.: Response calculations for light-scattering aerosol counters and photometers, Applied Optics, 4, 1463–1474, https://doi.org/10.1364/AO.4.001463, 1965.

Hogan, A. W., Barnard, S., and Bortiniak, J.: Physical properties of the aerosol at the South Pole, Geophysical Research Letters, 6, 845–848, https://doi.org/10.1029/GL006i011p00845, 1979.

IPCC: Technical Summary, p. 31–116, Cambridge University Press, https://doi.org/10.1017/CBO9781107415324.005, 2014.

Jaenicke, R., Dreiling, V., Lehmann, E., Koutsenoguii, P. K., and Stingl, J.: Condensation nuclei at the German Antarctic station “Georg von Neumayer”, Tellus B: Chemical and Physical Meteorology, 44, 311–317, https://doi.org/10.3402/tellusb.v44i4.15459, 1992.

Khlystov, A., Stanier, C., and Pandis, S. N.: An algorithm for combining electrical mobility and aerodynamic size distributions data when measuring ambient aerosol special issue of aerosol science and technology on findings from the fine particulate matter supersites program, Aerosol Science and Technology, 38, 229–238, https://doi.org/10.1080/02786820390229543, 2004.

König-Langlo, G., King, J. C., and Pettré, P.: Climatology of the three coastal Antarctic stations Dumont d’Urville, Neumayer, and Halley, Journal of Geophysical Research: Atmospheres, 103, 10 935–10 946, https://doi.org/10.1029/97JD00527, 1998.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmospheric Chemistry and Physics, 5, 715–737, https://doi.org/10.5194/acp-5-715-2005, 2005.

Massie, S. and Hervig, M.: HITRAN 2012 refractive indices, Journal of Quantitative Spectroscopy and Radiative Transfer, 130, 373 – 380, https://doi.org/https://doi.org/10.1016/j.jqsrt.2013.06.022, HITRAN2012 special issue, 2013.

Minikin, A., Legrand, M., Hall, J., Wagenbach, D., Kleefeld, C., Wolff, E., Pasteur, E. C., and Ducroz, F.: Sulfur-containing species (sulfate and methanesulfonate) in coastal Antarctic aerosol and precipitation, Journal of Geophysical Research: Atmospheres, 103, 10 975–10 990, https://doi.org/10.1029/98JD00249, 1998.

Palmer, K. F. and Williams, D.: Optical constants of sulfuric acid - Application to the clouds of Venus, Applied Optics, 14, 208–219, https://doi.org/10.1364/AO.14.000208, 1975.

Petzold, A. and Schönlinner, M.: Multi-angle absorption photometry—a new method for the measurement of aerosol light absorption and atmospheric black carbon, Journal of Aerosol Science, 35, 421 – 441, https://doi.org/https://doi.org/10.1016/j.jaerosci.2003.09.005, 2004.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X.-Y.: Recommendations for reporting "black carbon" measurements, Atmospheric Chemistry and Physics, 13, 8365–8379, https://doi.org/10.5194/acp-13-8365-2013, 2013.

Pratte, P., Cosandey, S., and Goujon-Ginglinger, C.: A scattering methodology for droplet sizing of e-cigarette aerosols, Inhalation Toxicology, 28, 537–545, https://doi.org/10.1080/08958378.2016.1224956, 2016.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle, Science, 294, 2119–2124, https://doi.org/10.1126/science.1064034, 2001.

Schwartz, S. E.: The Whitehouse effect – short-wave radiative forcing of climate by anthropogenic aerosols: An overview, Journal of Aerosol Science, 27, 359–382, https://doi.org/10.1016/0021-8502(95)00533-1, 1996.

Shepherd, R. H., King, M. D., Marks, A. A., Brough, N., and Ward, A. D.: Determination of the refractive index of insoluble organic extracts from atmospheric aerosol over the visible wavelength range using optical tweezers, Atmospheric Chemistry and Physics, 18, 5235–5252, https://doi.org/10.5194/acp-18-5235-2018, 2018.

Shettle, E. P. and Fenn, R. W.: Models for the aerosols of the lower atmosphere and the effects of humidity variations on their optical properties, Tech. rep., 1979.

Thompson, D. W. J., Solomon, S., Kushner, P. J., England, M. H., Grise, K. M., and Karoly, D. J.: Signatures of the Antarctic ozone hole in Southern Hemisphere surface climate change, Nature Geoscience, 4, pages 741—749, https://doi.org/10.1038/ngeo1296, 2011.

Toon, O. B., Pollack, J. B., and Khare, B. N.: The optical constants of several atmospheric aerosol species: Ammonium sulfate, aluminum oxide, and sodium chloride, Journal of Geophysical Research, 81, 5733–5748, https://doi.org/10.1029/JC081i033p05733, 1976.

Valenzuela, A., Reid, J. P., Bzdek, B. R., and Orr-Ewing, A. J.: Accuracy required in measurements of refractive index and hygroscopic response to reduce uncertainties in estimates of aerosol radiative forcing efficiency, Journal of Geophysical Research: Atmospheres, 123, 6469–6486, https://doi.org/10.1029/2018JD028365, 2018.

Virkkula, A., Koponen, I. K., Teinilä, K., Hillamo, R., Kerminen, V.-M., and Kulmala, M.: Effective real refractive index of dry aerosols in the Antarctic boundary layer, Geophysical Research Letters, 33, https://doi.org/10.1029/2005GL024602, 2006.

Wagenbach, D., Görlach, U., Moser, K., and Münnich, K. O.: Coastal Antarctic aerosol: the seasonal pattern of its chemical composition and radionuclide content, Tellus B: Chemical and Physical Meteorology, 40, 426–436, https://doi.org/10.3402/tellusb.v40i5.16010, 1988.

Weller, R., Wöltjen, J., Piel, C., Resenberg, R., Wagenbach, D., König-Langlo, G., and Kriews, M.: Seasonal variability of crustal and marine trace elements in the aerosol at Neumayer station, Antarctica, Tellus B: Chemical and Physical Meteorology, 60, 742–752, https://doi.org/10.1111/j.1600-0889.2008.00372.x, 2008.

Weller, R., Minikin, A., Wagenbach, D., and Dreiling, V.: Characterization of the inter-annual, seasonal, and diurnal variations of condensation particle concentrations at Neumayer, Antarctica, Atmospheric Chemistry and Physics, 11, 13 243–13 257, https://doi.org/10.5194/acp-11-13243-2011, 2011.

Weller, R., Minikin, A., Petzold, A., Wagenbach, D., and König-Langlo, G.: Characterization of long-term and seasonal variations of black carbon (BC) concentrations at Neumayer, Antarctica, Atmospheric Chemistry and Physics, 13, 1579–1590, https://doi.org/10.5194/acp-13-1579-2013, 2013.

Weller, R., Schmidt, K., Teinilä, K., and Hillamo, R.: Natural new particle formation at the coastal Antarctic site Neumayer, Atmospheric Chemistry and Physics, 15, 11 399–11 410, https://doi.org/10.5194/acp-15-11399-2015, 2015.

Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1 â€“ Evidence from measurements, Atmospheric Chemistry and Physics, 9, 3987–3997, https://doi.org/10.5194/acp-9-3987-2009, 2009.

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjäraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J. A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Grüning, C., Faloon, K., Beddows, D., Harrison, R., Monahan, C., Jennings, S. G., O'Dowd, C. D., Marinoni, A., Horn, H.-G., Keck, L., Jiang, J., Scheckman, J., McMurry, P. H., Deng, Z., Zhao, C. S., Moerman, M., Henzing, B., de Leeuw, G., Löschau, G., and Bastian, S.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmospheric Measurement Techniques, 5, 657–685, https://doi.org/10.5194/amt-5-657-2012, 2012.

World Meteorological Organisation, W.: WMO/GAW Aerosol Measurement Procedures, Guidelines and Recommendations, World Meteorological Organisation, WMO, 2016.

Zhang, X., Huang, Y., Rao, R., and Wang, Z.: Retrieval of effective complex refractive index from intensive measurements of characteristics of ambient aerosols in the boundary layer, Opt. Express, 21, 17 849–17 862, https://doi.org/10.1364/OE.21.017849, 2013.

Zieger, P., Aalto, P. P., Aaltonen, V., Äijälä, M., Backman, J., Hong, J., Komppula, M., Krejci, R., Laborde, M., Lampilahti, J., de Leeuw, G., Pfüller, A., Rosati, B., Tesche, M., Tunved, P., Väänänen, R., and Petäjä, T.: Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study, Atmospheric Chemistry and Physics, 15, 7247–7267, https://doi.org/10.5194/acp-15-7247-2015, 2015.