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Atmospheric Chemistry and Physics Discussions

Interactive comment on "One year of aerosol refractive index measurement from a coastal Antarctic site" *by* Z. Jurányi and R. Weller

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

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. We are not really 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. Hygroscopicity measurements are planned for this austral summer (winter 2019-2020) at our measurement site which will hopefully 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 was done to the manuscript.

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 reformulated, 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, it was removed 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 RIeff.

answer: The standard deviation value of RIeff 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: The paragraph was rewritten: "We find no significant dependence of the derived RIeff values on the wind direction. Thus, we conclude that RIeff 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 RIeff. Just in one case, a temporary high contamination episode with diesel engines operating right next to the measurement site resulted in an unusual high RIeff 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 con-

fusing 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: The sentence was removed 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: This additional text was added to the abstract: "To conclude, our study revealed largely constant RIeff values throughout the year without any sign of seasonality. Therefore, it seems reasonable to use a single, constant RIeff 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 wright, "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: Was removed.

reviewer comment: Page 2, line 34: What does it mean, “Until the particles disap-

peared”?

answer: The text was changed 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: To dry aerosol, the line was modified 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: “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: The sentence was modified 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. A definition to RH and the following sentence was added 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. The explanation was added to the text and it was renamed 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: done, the sentence now reads: "This strong seasonal variability is the reason why in summer a much shorter time averaging period is sufficient to enable a successful RIeff 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 RIeff fit was possible, was added 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, at least one hour but up to 14 hours. But that is not the problem. The problem is if the aerosol changes significantly within one scan, which lasted 6 minutes for both instruments. 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 time period. The SMPS on the other hand scans through the different diameters during 6 minutes and therefore captures only one size at a time. Therefore e.g. when during the 6 minutes of a single scan the aerosol concentration doubles and the form of it remains constant, the LAS returns the average number size distribution, whereas the SMPS returns a skewed number size distribution with too low concentrations at the low diameters and too high at the high diameters (assuming that the SMPS is operated in an upscan mode). And yes, it could be also seen in the CPC data, that during the beginning of the scan the concentration was much higher than later. The text was changed 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 ($\sim$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: done

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

answer: it was determined by a statistical T-test with a significance level of 0.01. The 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: the standard deviations are added now.

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 showed that that the WSOC mass fraction 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. This was added to the text as well: " 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-SO42-/NH4+ 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 ((NH4)2SO4) is more plausible. However, given the high nss-SO42- excess (corresponding to a high H2SO4 excess), partly ammoniumbisulphate (NH4HSO4) may also be formed. The RI of NH4HSO4 is lower (ChÃ¡lek et al., 1995) than that of NH4NO3 (1.473 instead of 1.52), but the latter would be comparable to that of (NH4)2SO4 (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: The figure was expanded. 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, no obvious contamination could be identified among these scans. Such scans when there was a known contamination present, such as the scan in figure 8, was removed from the data analysis.

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: Units are provided. The scattering cross section is a well-known physical property, it 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. We included

the method of calculation now in the text as well (our 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. The sentence of Line 14 states it: "Figure 9 shows the time average of dsigma/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. It was also added to the 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. Now, the figure caption states as well, that the data is an average.

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

answer: The sentence: "The resulted RI values will describe the particles with the particle sizes of the corresponding size range." was changed 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 lines meet around log(D)=3 which is D=1000nm. Our highest diameter range for the size dependency investigation was 478-710nm. Even at the largest diameter of 710nm (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 even at our highest diameter range is meaningful. 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: the geographic border was used to express, to question where exactly can a single (season independent) RI value of 1.44 be used. Is it only our measurement site? Or is it also valid 50 km away? May it be valid for other coastal Antarctic sites? Or maybe even everywhere in Antarctica?

The text was changed 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 RIeff 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: The standard deviation of the RIeff and the chemical composition RI is now

included 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: The values were provided.

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. âĂČ

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