

Atmos. Chem. Phys. Discuss., referee comment RC2  
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## **Comment on acp-2020-1311**

Anonymous Referee #2

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Referee comment on "Seasonality of the particle number concentration and size distribution: a global analysis retrieved from the network of Global Atmosphere Watch (GAW) near-surface observatories" by Clémence Rose et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1311-RC2>, 2021

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My apologies to all for the tardy review. I view the data analyses in this paper as strong, but some of the interpretations and conclusions derived from the analysis are not nearly so rigorous as the methods. The introduction is nicely written, and the analysis of the fraction of available data that is important to adequately represent the statistics appears to be very well done. At about 31 pages of text and 13 figures, the paper is too long.

Section 5.2 is a series of cursory discussions about what the variations in  $N_{tot}$  may represent. The discussion here does not offer a lot, and some of the inferences concerning POLAR sites are a little misleading. It might be helpful if the authors told us how these data should be used to help global models: e.g., which plots might you think the most useful for model evaluation?

Detailed comments

- Page 2, lines 43-44 – It is unclear whether multiple transformations include wet and

dry deposition. Explicitly mention deposition.

- Page 3, line 10 – maybe “less evident” rather than “barely marked”.
  
- Page 3, line 18 – “>50-100 nm” is unclear. Instead, maybe “(considered here as either >50 nm or >100 nm)”.
  
- Page 5, lines 17-20 – The wording here suggests that such closure studies have never been conducted until recently, which is not the case. Please add something like “at the above-mentioned network sites”. The ACTRIS reference relates to the Schmale study, not this generalized statement.
  
- Page 5, lines 31-33 – It is very disappointing to see no PSND data from Alert and no data at all from the Whistler Mountain site (both of which have many related publications) not included in the “most up-to-date information ... worldwide...”.
  
- Figure 1a – “Pristine” is defined as “belonging to the earliest period or state”, “not spoiled, corrupted, or polluted (as by civilization)”, “fresh and clean as or as if new”. In terms of the aerosol, this definition means that a site is pristine if it is not influenced by anthropogenic sources, rather only by natural sources. Parts of the Arctic can reach a pristine condition during the summer, but it is incorrect to broadly represent the Arctic

stations as pristine. Arctic Haze is not a pristine condition. The general use of 'pristine' to describe the polar sites is unjustified.

- Section 3.2 – You state that you fit two modes to the particles between 20 nm and 500 nm, and the first mode is comprised of Aitken particles (maybe nucleation particles, depending on one's definition). Is there a typical size range for the minimum between the two modes, and can this minimum be related in some cases to cloud processing (i.e., Hoppel minimum)? Also, modes of some urban and rural distributions can be singular. Is that allowed in your analyses? Lastly, is 100 nm your definition of the split between Aitken and accumulation, and would you be explicit about that, please.
  
- Section 3.3 – Two points are missing here: 1) CCN number concentrations do not necessarily translate to cloud droplet number concentrations, particularly at higher concentrations, due to competition for water vapour by droplets growing at cloud base; 2) particles at 50 nm can only be effective if the cloud base supersaturation reaches a sufficiently high value. The latter happens in one of two ways: relatively few particles larger than 200 nm, such that the rate of water uptake by the >200 nm particles does not dominant the total uptake; very rapid cooling rates, typically associated with relatively extreme updraft speeds.
  
- Page 16, lines 18-30 - There are others in Figure 2 that appear to fall into the category of 95% or more: ZEP and ALT, LLN, WGG. Why were they not used? Also, I find it odd that data coverage can be essentially 100%, as it appears to be for TRL and NMY. Do these sites not perform checks on the operation of the instrument, zeroes, etc.? The filtering for wind direction, referred to for SPO, BRW and MLO, also happens for the ALT data, and it has been shown that that such filtering of the Alert data has little impact on the observations at Alert, except for a near-negligible impact on Aitken-size particles.



- Page 24, lines 19-20 – The Arctic and Antarctic are quite different in some respects, most notably the strong influence of southern-latitude anthropogenic sources on the Arctic in the winter and spring, also known as Arctic Haze. Arctic Haze is significant, and the number concentrations of accumulation-mode particles can extend through significant depths of the Arctic troposphere. Your generalized statement, based on measurements at the Antarctic, should not include the Arctic.
  
- Page 24, lines 28-29 – I don't understand the statement beginning "Larger diameters ... during JJA". One of the reasons that  $N_{tot}$  is higher at Arctic sites in the summer (JJA) is because the accumulation mode concentrations are much lower, which helps NPF (e.g., Freud et al., ACP, 2017; Croft et al., ACP, 2016; Leaitch et al., Elementa, 2013). This statement appears to say the opposite.
  
- Page 24, lines 32-33 – "Transport..." – Yes, but for accumulation-mode particles the transport in Croft et al is accompanied by stronger wet deposition, which significantly reduces those particles.
  
- Page 25, lines 6-8 - Arctic Haze, which has been studied for over 50 years, is pervasive during the winter and spring in the high Arctic, largely because of the reduced deposition of these particles in the Arctic during those times of the year. Your statement makes Arctic Haze sound like an occasional factor, which it is not.
  
- Page 28, lines 13-14 – It may be fair to say that the higher  $N_{tot}$  at mountain sites





- Page 40-41, lines 34-4 – You overlook the issue with the impact of particle concentrations on cloud supersaturation, which is just as important as the numbers of CCN.

- Page 44, lines 4-6 - Figure 5a indicates relatively uniform global coverage, although, as you mention, Africa, Russia, South America and oceans are clearly under-represented. Do you need all 39 European sites? Why not introduce a bit more global balance by conducting the analysis using only the sites in Figure 5a plus a smaller selection of European sites?

- Page 44, lines 22-23 – It may be true for some of the forest sites, mountain sites and polar sites, but it does not appear to be true for the urban or rural background sites.

- Page 45, lines 2-3 – Based on section 3.2, it seems as if your analysis essentially dictated the bimodality. Is that false?

- Page 46, line 10 – Remove "Slight".