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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the seasonal variation of observed size distributions in Northern Europe and their changes with decreasing anthropogenic emissions in Europe: climatology and trend analysis based on 17 years data from Aspvreten, Sweden" *by* Peter Tunved and Johan Ström

Anonymous Referee #2

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Review of "On the seasonal variation of observed size distributions in Northern Europe and their changes with decreasing anthropogenic emissions in Europe: climatology and trend analysis based on 17 years data from Aspvreten, Sweden", by Peter Tunved and Johan Stroem. (submitted to Atmos. Chem. Phys.).

This manuscript presents an analysis of 17 years of near-surface aerosol particle size distribution measurements at the Aspreveten regional background monitoring site in

Central/Southern Sweden. The topic of the research is of particular interest to understand how the tropoapheric aerosol layer has responded to reductions in emissions that have occurred in the 1980s and 1990s, re: subsequent climate influences from aerosol-radiation-interaction and aerosol-cloud-interaction radiative effects.

The paper represents an interesting and novel study of the 17 years of size distribution measurements, with trends applied on different types of size distribution, identified via cluster analysis. The approach may represent an important advance, the technique complimenting other studies size-resolved aerosol trend analysis studies, stratifying into size-distribution-types then enabling to assess process-linked changes, with then potential to explore for any signals of tropospheric aerosol layer response to reduced emissions in recent decadaes.

However, the Abstract and Introduction in the current version of the manuscript require substantial improvement and the initial summary statistics will be confusing to some readers as presently worded. The authors need to improve the explanation of these initial overview comparison plots to put the Asprevreten site, and the occurrence of new particle formation events, into better context in comparison to other size distribution monitoring sites in Scandinavia: Hyytiala, Pallas, Varrio and Vavihill.

It remains surprising to me that the seasonal average size distributions in Figure 2 seemed to suggest only two sub-micron modes – an Aitken mode at about 50-80nm diameter and an accumulation mode at about 200nm diameter, and yet the statistics from log-normal fits to the hourly measurements in Table 1 identify three modes – with a distinct nucleation mode at 30-40nm, in addition to an Aitken mode at 60-70nm.

The daily-mean size distribution overview plot in Figure 1 shows a clear seasonal variation in the aerosol measured at Aspreveten, total number concentrations increasing from 1000 per cc in winter to about 2000 per cc in summer, with Aitken mode peak at Dp=50nm in winter, increasing to 100nm in summer. And at least from these daily-mean plots in Figures 1 and 2, there is no obvious sign of a nucleation mode, whereas

in Table 2 the nucleation mode has approximately equal particle concentrations to the Aitken mode.

The manuscript needs to explain the reason for this apparent discrepancy, and also put the Aspreveten site analysed here into better context with the other Scandinavian sites.

Strong diurnal variation occurs during new particle formation (NPF) events (e.g. as in Figure 2 of Kulmala et al., 2004), from initial NPF early-morning and subsequent growth through the morning and early-afternoon, the air measured through the day then having different times from growth then manifesting as the so-called banana plots in dN/dlogr vs time within the air sampled at the monitoring site. And it is likely the case then that these NPF variations will be less apparent within daily-mean or daily-integrated aerosol measurements. However, the difference in these statistics may be underlining the importance of applying the trends analysis on the cluster-analysis-stratified data, compared to what is apparent from more daily-mean data, which will average out much of these important variations.

Is it the case that Aspreveten has weaker or less frequent nucleation days than Hyytiala and other near-forest sites, perhaps due to much less biogenic VOC emissions from forests, and subsequent influence on new particle formation (e.g. Metzger et al., 2010)? Or is it simply that the nucleation events are occurring, but daily-means smooth out the strong diurnal variation in NPF events?

If it is the latter, there may be important implications for the way global modelers design model experiments, I mean in terms of diagnostics to retain these important variations. Often modellers are ambitious in attempting to be comprehensive, to compare measurements to a large number of datasets, and likely consider a compromise necessary in choices of diagnostic information to include in the model experiments. However, in the case of new particle formation it may be a pre-requisite to ensure information on the variability on an hourly timescale is retained (perhaps with some approach to store the

mean of the square-of-a-metric as well as the mean of a metric to enable the variability to be reconstructed, with some optimal sampling and/or re-initialisation sequence).

I am not suggesting the authors make this specific point, but, if the statistical variation is indeed the reason for the apparent discrepancy, to consider adding a more general note, either in the discussion or conclusion-bullet-points about any re: how modellers can efficiently/effectively retain the diurnal variation information in their simulations. For example a statement along the lines of the hourly-mean and daily-mean statistics here underline the importance to consider how best to design diagnostics in models to retain information on new particle formation within co-ordinated composition-climate model experiments.

My other major concern is re: section 4.4 where the CALM model from Tunved et al. (2010) is applied, and some additional calculations are made to estimate the potential cloud albedo change of the trend period. I could not follow the calculations on page 15 of the manuscript, and do not see a reference for the application of the CALM model for this application. I suggest the authors consider whether it might be best to restrict the scope of the article to describing the aerosol changes.

It is my opinion that, with some additional revisions (which I classifiy here as major, but may actually be relatively straightforward to make) some important conclusions can be identified from combining the cluster analysis and the 17-year trend analysis. My recommendation is the authors restrict the scope of this paper to the aerosol changes and consider whether co-operation from others might enable the approach to potentially be applied also on other sites with long-term trends (e.g. those in Asmi et al., 2013)? In that scenario, the article could well stand independently from section 4.4, the analysis potentially benefitting also from a more focussed scope on this single site, and also identifying a potential future article.

To summarise, my review is to recommend major revisions, including re-drawing Figure 1 adding in an extra sub-panel (Figure 1b, see point 30 below) to present the variability

within the daily-means – perhaps simply the ratio of the standard deviation to the mean would provide, via the coefficient of variation, an indication of which parts of the particle size range have substantial variability, likely highlighting days where new particle formation and growth is occurring upwind of the site. I think this could be a good way to provide overview analysis of the seasonal occurence of these events, and link then to explain the discrepancy with the hourly-mean statistics in Table 1.

I have restricted the scope of this review mostly to the Abstract, Introduction and initial Figures – the above issues seeming to me to require a further substantial revision to the manuscript, but can confirm I am willing to review the revised manuscript once the specific revisions I recommend below have been made.

Specific revisions ——————

1) Abstract, line 9 – add "at a rural background site in Northern Europe" (or similar) after "trends", and corrected grammatical error "investigate" –> "investigated".

2) Abstract, line 13 – delete "has been" and be more specific when you say "during last decades" – maybe it's just a case of replacing with "during the 17-year trend period"? Also, I think one of the key things in your findings is that your analysis identifies that particle size distribution has shifted as well as the particle number. The previous N20 and N100 trends may also show this to some extent, but the size-resolved cluster analysis at this single site here may be better able 17 to demonstrate this? So you could maybe strengthen the point about particle size by "We show that, not only have particle number concentrations decreased, but also particle size has shifted, with potential implications for aerosol-climate influence?" You could also add something to the conclusions re: future work to further investigate this at other sites?

3) Abstract, lines 14-15 – the 2 sentences here should be joined into a single sentence, so that then the two sentences following on from the previous point are first one re: changes in particle number, and then the 2nd one re: the shift in particle size? The wording just needs to be improved, if possible.

4) Abstract, lines 17-18 – this setence again should be strengthened to make the point that the shift in size affects mass, but add also re: the importance then for radiative effects.

5) Abstract, lines 19-22 – rather than stating "a rather complex picture emerges" these sentences here need to summarise specifically the seasons where the main decreasing trend is seen – then point out which seasons or modes show the increases (is it just, Aitken mode and summer?)

6) Abstract, lines 22-24 – all previous text has referred specifically to the measurements at Aspreveten, but this sentence is (I think) referring to the trajectory model analysis you've carried out? If that's correct then rather than "data analysis", instead give additional/different words to explain that it is from simulations with the trajectory that this finding is demonstrated? Or if it is simply your interpretation of this, then state "We interpret this as.. " or similar. Also – re: the "receptor" – here you mean the actual Aspreveten site is a receptor – maybe better to explain this as "receptor site" or "upwind of the receptor site"?

7) Abstract, lines 24-26 – is "an adiabatic cloud parcel model" the right description for the analysis tool applied for section 4.4? The CALM model described in Tunved et al. (2010) seems to be predicting the evolution of the size-resolved aerosol with some influence from clouds. The simulations can predict the CCN considering air-parcel trajectories, but it is, to my understanding, not really a cloud parcel model. Please re-word this sentence, and the opening paragraph to section 4.4 accordingly.

8) Abstract, lines 26-27 – as per my main comments above, I am recommending the authors restrict the scope of this article to the aerosol changes, in which case this additional finding re: the 10-12% cloud albedo change may not need to be included.

9) Introduction, page 2, lines 2-4 – the choice of "Turbidity" in this opening sentence was unusual and is, in my opinion, a good alternative word to describe the optical thickness of the atmosphere. The word was often used in the articles in the 1960s

and 1970s, for example re: the dimming effect from the volcanic aerosol haze after the 1963 Agung eruption (e.g. Volz et al., 1970), but these days is seldom used in journal articles. The original 1908 article by Gustav Mie used this term to describe the aerosol-radiation interaction effect (see e.g. Lilienfeld et al., 1991) and it is, in my opinion, good terminology. However, in the 2nd sentence, with the Mie scattering from aerosol, and the associated radiative effects from aerosol changes these days a core requirement within all climate model integrations, suggest to refer to either a review article and/or relevant text book (e.g. Seinfeld and Pandis, 1998) when introducing these basic effects for explaining the solar dimming and the relationship to the size-resolved aerosol. Also, in addition to explaining a correspondence with aerosol volume in the accumulation mode, suggest to add these days the understanding of the origin of such particles being from sizes as small as a few nm, and to understand the importance of new particle formation (e.g. cite Kulmala et al., 2004 for the measurements that have demonstrated this).

10) Introduction, page 2, line 9 – Please follow the recommended way to cite IPCC climate assessment reports, via citing the relevant chapter, e.g. Mhyre et al. (2013) for the radiative forcing chapter.

11) Introduction, page 2, line 10 – suggest to change "indicate that dimming" to "show that overall solar dimming"

12) Introduction, page 2, lines 11-13 – re-word this sentence to more clearly explain the brightening trend – it is not clear what you mean by "effect of economic growth" – I think you mean economic growth in East and South Asia, with the 2nd part of that sentence on air quality legislation being re: emissions reductions Europe and North America. The overall brightening effect suggests the aerosol decreasing overall, and you need to clarify this sentence to explain this better, perhaps citing other articles such as Ohmura (2009). Although the next paragraph indicates the regional changes, this initial sentence needs to be clearer.

13) Introduction, page 2, lines 15-16 – add "vapour" after "sulfuric acid" in both instances here, so be clearer re: the subsequent condensation onto particles and driver of new particle formation. Suggest also replace "sulfate aerosol mass" with "sulfate aerosol particles" or "sulfate aerosol particle mass" so it's clear you mean the aerosol particle phase. Suggest to change "condensation onto already existing particles" with "condensation, which grows existing particles".

14) Introduction – page 2, line 19 – re-word "with the exception perhaps of the Indian sub-continent". Although there are uncertainties you can be more certain about the different trends in emissions and suggest to describe the regions more regionally such as South Asia, citing a recent paper on this, and perhaps draw a distinction between aerosol precursor emissions and observed changes re: other emissions such as carbonaceous aerosol and influences from other precursor emissions or oxidation processes.

15) Introduction – page 2, line 23 – the term AOD refers to integrated aerosol extinction, and hence just the "aerosol optical depth" not "atmospheric optical depth". Please correct this.

16) Introduction – page 2, line 25 – this seems too detailed information on North America, and more detail on any differences in trends within Europe would be better here – does the literature contrast trends in different parts of Europe (e.g. contrast Northern Europe to those observed in Central Europe and in Southern Europe?).

17) Introduction – page 2, line 29 – suggest to delete the trends in Arctic and Antarctic to give room for the above suggested discussion re: any literature on different aerosol trends within Europe.

18) Introduction – page 2, line 30-34 – reduce this para on Turnock et al. to focus on the main findings, again add some additional discussion re: aerosol changes in different regions of Europe.

19) Introduction – page 3, lines 10-12 – reword "However, due to claimed lack of data" – to explain instead as sites with more than 10 years of data for the trend analysis. Also there were 5 sites in that study, not 4 – Melpitz in Germany was also included in the analysis.

20) Introduction – page 3, lines 23 – reword "Clearly there is lack of long-term aerosol microphysical data" to be more positive. These days, following the programs to establish additional sites to monitor aerosol size distribution, there are several additional sites that have 10 years of data, for example the EUSAAR sites analysed in Asmi et al. (2011), with SMPS/DMPS aerosol measurements since the time of the EUCAARI EU FP6 integrated program (Kulmala et al., 2009).

21) Introduction – page 3, lines 27-30 – reword this sentence maybe to be clearer you mean these monitoring sites are providing valuable new understanding of aerosols, and in particular trends in Northern Europe. Maybe the start of that sentence add "in Scandinavia" (rather than later) or "in Northern Europe", e.g. re: where 4 of the 5 sites in Asmi et al. (2013) analysis are located.

22) Introduction – page 4, line 16 – "The explanation for this" – cite a paper that has already shown or explained this effect – perhaps cite Kerminen and Kulmala (2003) or other paper re: the role of the condensation sink and new particle formation?

23) Introduction – page 4, line 19 – "Based on the literature referenced above there is a notion" Re-word this to be clearer citing relevant paper etc. I think you can be clearer to state this is well-established to be the case. There is a general question of the "aerosol response" to the reduced SO2 emissions in different parts of Europe and perhaps this part of the Introduction you could bring the earlier literature review round to bigger-picture question and then explain the specific analysis this paper presents.

24) Methods – page 5, lines 19-20 – order the 2003, 2004, 2005 papers chronologically here.

25) Results – page 8, line 32 – explain a bit more re: the link between the lack of photochemistry and the absence of small particles – you could maybe state the oxidation of SO2 specifically here and the short atmospheric residence time of the sulphuric acid vapour, or so.

26) Summary and conclusions – page 19, lines 1-4 Replace "increasing on expense of" with "with a reduction in" and replace "vanished during the period 2000-2017" with "reduced to xx% at the end of the trend period" or similar.

27) Summary and conclusions – page 19, lines 23-26 The text for this bullet begins stating no evidence for increased nucleation was found. But then the last sentence says there was more frequent nucleation upstream of the site. Please clarify – as per earlier comments, I think the finding of increased nucleation is important one and perhaps the revised paper could focus more on identifying this and adding re: future work to understanding the overall aerosol reponse in other sites and regions of Europe?

28) Summary and conclusions – page 19, lines 29-32 – and page 20, lines 1-5 As per my main comments, I wonder whether best to restrict the analysis to the aerosol trends rather than the activation and cloud albedo calculations? These final 2 bullet points (particularly the last one) do not add substantial findings – suggest better to focus on the main conclusions re: the aerosol trends.

29) References – the References section seems to have reverted back to a different font. Please remedy this to match the required style for ACP.

30) Figure 1, page 24 – as per my main comments, I was trying to understand why the daily-mean size distributions seem (from this initial Figure) to suggest only one mode across the nucleation and Aitken, whereas the fits to the hourly measurements in Table 1 then reveal the presence of the distinct nucleation mode. I'm suggesting here whether showing as an extra panel b) in this Figure you could show the relative variability within the 24 hours of data (for each size bin) – maybe plot the ratio of the standard deviation to the mean – i.e. the coefficient of variation – to show which parts

of the size range then have most variability – perhaps this will then identify this variation and reason why the nucleation and Aitken modes in Table 1 are only apparent in the hourly data?

31) Figure 2, page 25 – the presence of the accumulation mode in these seasonal size distribution pdfs is apparent on this linear y axis, but it would be clearer if the plot showed these size distributions on a log-y axis. This also relates to the trends in particle number, with a number concentration change of 10 per cc being more significant for the accumulation mode than for the Aitken or nucleation modes. Suggest to re-plot this Figure with the dN/dlogr axis showing then 10 to 10,000 per cc or so, to be able to see the variations in number in the accumulation mode, as well as those in the Aitken mode.

32) Table 1, page 26 – the median and inter-quartile range for the nuclei and Aitken modes are similar, from these hourly size distribution log-normal fits, but the presence of the 2 modes is not apparent in the daily-mean size distributions in Figures 1 and 2. See main comments, and points 30 and 31.

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