Dear Editor, der Referees!

We thank you for your efforts. Below are our answers to the points that were raised in the second round of this review process. We hope that we can answer them to your satisfaction. The reviewer comments are given in blue, our answers are given in black. In the updated version of the manuscript, changes are indicated in bold script.

Please let us know if this suffices for you to accept our manuscript for publication in ACP.

Sincerely,

Heike Wex

Referee #3

Figure 9b shows only the differences of N_{CN}. So it should be good to show not only the differences of N_{CN} but also the differences of N_{CCN} at several supersaturation values between CEs and MEs (perhaps excluding precipitation days in MEs) in Table 2. Since difference of aerosol concentration between CEs and MEs is depend on aerosol size, adding representative percentile concentration of CCN would be informative.

We made a new Figure 9 showing the scatter plots and box and whisker plots additionally for CCN for two supersaturations (0.3% and 0.7%) exemplarily, and adjusted the text accordingly (see page 12, line 15ff). As respective values had been given before (and are still given) in Table 2, no additional new discussion was needed.

Figure 10: N_{CCN} at different supersaturations did not seem to show time lag. Can the authors insist that the nucleated particles grew to a size that could act as CCN as time progressed? Concentration of large particles (SS=0.1%) also tended to increase with time.

Concerning an observed time lag, we now say “accompanied” instead of “followed” (page 13, line 12), as this indeed captures all cases (it’s only for some cases that the increase in size in larger particles started delayed).

Concerning the increase in concentrations of large particles, we thank you for noticing that. The absolute changes are quite small, but you are correct. We therefore changed this passage by deleting original lines 12 and 13 on page 13 and changed the remaining text (now page 13, lines 13-14) to:

“In the vast majority these events of increased N_{CN} were accompanied by an increase of N_{CCN} by a factor of at least roughly two at all SS.”

Nevertheless, in general, we clearly see newly formed particles, indicated by a strong increase particularly in small particles (~ > 35 nm) and also an increase (often ~doubling) in number concentrations for particles with sizes of roughly 35, 45, 60, 80 and 125 nm (these sizes are rough estimates based on an assumed kappa value of 0.8 and on the supersaturations in the CCNc, and particles of particularly the larger sizes surely can act as CCN). If this increase in concentration at the

larger sizes would not come from newly formed and condensed particulate matter, another explanation would have to be found for the observed increase. The authors are not aware of an alternative useful explanation of the observation. If one is known we are open to discuss it. For the time being, concerning this topic, nothing was changed.

Figure 11: Add the PSCF results for $N_{CN > 90\ nm}$ (LAS). I think this can be compared to the result of $N_{CCN,07}$ and $N_{CCN,01}$.

As can be seen in Figs. 6-8 (panel C in all cases), $N_{CN > 90\ nm}$ (LAS) and N_{CCN} at 0.1% supersaturation ($N_{CCN,01}$) are very close to each other (symbols in red and darkest blue) throughout the whole time when measurements were made. The PSCF result for $N_{CN > 90\ nm}$ (LAS) is therefore very similar to that show in Fig. 11, lower right panel (for $N_{CCN,01}$). Adding it does not add anything new to the work and only unnecessarily makes it longer. Nothing was changed.

Referee #1

One point I keep wondering about is in how far the PSCF results reflect the source region of precursor gases of secondary aerosol formation. As is now discussed in the work, NPF can occur in the free troposphere, so the location of the formation can be understood as source region. This source region would, however, not reveal where the precursor gases came from because they can travel over long distances in the atmosphere before a change in CN or CCN can be observed (e.g., Woodhouse et al., 2013). It is beyond the scope of this paper to investigate this systematically, but a short mentioning of this would clarify the type of information that can be extracted from the chosen methodological approach. My recommendation is to publish the manuscript with this minor addition.

It is correct that the PSCF does not exactly show where precursor gases came from, NOR does it show where the new particle formation takes place. We added the following (page 14, line 29ff):

“While with this interpretation we assume the source regions to be the regions where NPF may have taken place, gaseous precursors may have been emitted in these regions, too, or may have been transported over some distance. Still, our analysis clearly indicates that the Southern Ocean region is a region potentially acting as a source of the majority of particles observed at PES.”

We did, however, not find a way to incorporate the citation you gave. As we understand this cited work, the outcome concerning the Southern Ocean is rather that changes in DMS emissions there will not have a large influence on CCN number concentrations. The statement made above “the precursor gases … can travel over long distances in the atmosphere before a change in CN or CCN can be observed (e.g., Woodhouse et al., 2013).” is, to our understanding, not made in it, unless you refer to what could be envisioned in their Fig. 3b, which, however, is not discussed in their text.

Literature

Woodhouse, M. T., Mann, G. W., Carslaw, K. S., and Boucher, O.: Sensitivity of cloud condensation nuclei to regional changes in dimethyl-sulphide emissions, Atmos. Chem. Phys., 13, 2723-2733, 2013.