Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2020-1202

Anonymous Referee #1

Referee comment on "Wintertime subarctic new particle formation from Kola Peninsula sulfur emissions" by Mikko Sipilä et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1202-RC2, 2021

General comment

The manuscript provides an interesting perspective on the effect of anthropogenic emissions for wintertime new particle formation (NPF) in the Arctic. In particular, Sipilä et al. show that the high SO2 emissions from industrial activities in the Kola peninsula (Russia) can lead to sufficiently high sulfuric acid concentrations to promote NPF despite the low amount of solar radiation. Previous studies[1] have already linked high level of SO2 from the Kola peninsula with NPF occurrence but this is the first study that quantifies sulfuric acid concentration and provides a molecular level characterization of the nucleating clusters.

The analysis is generally sound and the results are nicely presented. However, the manuscript focuses too much on a single case study. A more comprehensive analysis of the entire campaign would be highly beneficial to better understand the impact of industrial emissions on NPF compared to other processes. The authors, could for example look at the effect of SO2/H2SO4 and/or wind direction on the occurrence of NPF events. I also recommend to include a table listing all NPF events with the most relevant variables (e.g. wind direction, H2SO4 and SO2 concentration, J rate if available), this could be a very useful reference for future studies.

The manuscript would also benefit from a careful proofreading.

Specific comments

Nucleation rate calculation: I am concerned about the application of Stolzenburg[2] equation to estimate GR2. This equation was developed for the growth of neutral particles

but you are using it for ions. The growth of charged nano-particles can be significantly faster because of the dipole moments of sulfuric acid [2, 3]. I understand your concern about fitting the entire PSD as shown in Fig.S1, which can lead to an overestimation of the particle growth rate due to airmass advection. A possible solution could be to apply the appearance time method to the growth of ions smaller than 3nm. The lifetime of these ions is comparable to sulfuric acid and I think they should not be affected much by air mass advection. The advantage of this method would also be that it does not rely on the assumption that sulfuric acid condensation is the sole responsible for growth.

Ion induced nucleation (IIN): you often refer to IIN as the driving NPF mechanism. However, without a measurement of neutral particle formation rate is not really possible to say if IIN is the dominant mechanism. I agree that, based on previous experiments/field observations, IIN would probably play a major role given the low sulfuric acid concentrations. However, you should state more clearly and earlier in the text that this is just an hypothesis. Moreover, I am also wondering to which extent the low ion formation rates can explain the increase in the particle number for the event presented in the main text. In particular, you report a maximum formation rate of about 6E-2 ions/(cm3*s) (Fig.5) but the increase of particles larger than 3nm shown in Fig.7 is larger than 1E3 over the course of an hour roughly, which translates in a formation rate of about 0.3 particles/(cm3*s). This rate is 5 times faster compared to the ion formation rate and it is just a lower limit estimate (losses are not considered). Do you have an explanation for this? Maybe it would be beneficial to compare the ion formation rate with the formation of neutral particles larger than 3nm.

Sulfuric acid proxy: I appreciated the application of the sulfuric acid proxy from Dada et al. 2020[4] to estimate the sulfuric acid concentration and it is nice to see the agreement on Fig.5. However, I would like to see a comparison between the sulfuric acid measurement and the proxy for the entire campaign when data are available. This comparison would be helpful to understand the applicability of this proxy to other Arctic locations where sulfuric acid measurements may not be available, providing a useful reference for the entire community. I was also intrigued by the hypothesis that the discrepancy between the proxy and the measurement on January 28 may be due to the strong surface inversion and it would be interesting to see how often this effect is present. If you do the comparison it should be easy to see if stronger surface inversions lead to higher discrepancies.

Particle growth: in the manuscript you show that that particle growth cannot be explained by species measured with the nitrate CIMS (i.e. sulfuric acid, MSA, iodic acid and HOMS) and conclude that the growth must be happening closer to the source. I think this hypothesis is plausible, however, you should mention that the growth could also be driven by condensation of organics which are not detected by the nitrate CIMS. Stolzenburg et al. 2018[5] have shown very clearly that at -25C nano-particle growth is mainly driven by organics not detectable with a nitrate CIMS. I can imagine that the industrial activities in the Kola peninsula also lead to high organic emissions and these may very likely contribute to particle growth. Unfortunately with your dataset is not possible to distinguish between these two possibilities but you should discuss both of them and probably leave the question on the growth mechanism open.

Minor comments
Line 12: I would mention the first source of air pollution in the Arctic
Line 30: OECD is not defined.
Line 30: "converged" you mean decreased?
Line 46: you should mention that SO2 leads to sulphate accumulation in aerosol particles as well (most of the clouds do not precipitate).
Lines 72-83: Include also nitric acid nucleation
Line 103: "another DMPS"? You mean one of the two DMPS?
Line 127: What is the frame size? Maybe you can remove this info as it is not very relevant
Eq. 2: the notation with Dp/nm is confusing, I would use Dp [nm]
Line 198: What does a "reasonably strong event" mean?
Line 233: there is a reference missing probably.
Lines 321-335: I like the application of Schobersberger et al. 2015[6] results but you should mention that fragmentation can have an effect on this comparison.
Line 348: Frege et al.[7] already reported the observations of HIO3-H2SO4 clusters, so I would not say that this has not been observed before.
Line 368: Something is missing in the sentence.
Line 386: Do not start a sentence with >100
Line 399: The acidity of aerosol particles is not necessarily related with their hygroscopicity and anyway you don't have information on particle composition so I would just remove this sentence.
Data availability: the link does not work and as stated by the ACP data policy all data should be deposited in a public repository. Please avoid using the "data are available upon request" sentence.
The reference to Dada et al. 2020 in the bibliography is missing.

Figure 2: You should not use the Jet color map, can you switch to a perceptually uniform colormap (e.g. parula in matlab or viridis in python)?
Figure 4: The figure is not easy to read, for example there are not relevant information and some trajectories have the same color. Can you replot the Hysplit data instead of just copying the original output?
Figure 5: The ion color scale is missing, also change the jet color map.
Figure 7: Change the jet color map.

References

References
[1] Kyrö et al. 2014 https://doi.org/10.5194/acp-14-4383-2014
[2] Stolzenburg et al., 2020 https://doi.org/10.5194/acp-20-7359-2020
[3] Svensmark et al., 2017 https://doi.org/10.1038/s41467-017-02082-2
[4] Dada et al., 2020 https://doi.org/10.5194/acp-20-11747-2020
[5] Stolzenburg et al., 2018 https://doi.org/10.1073/pnas.1807604115
[6] Schobersberger et al. 2015 https://doi.org/10.5194/acp-15-55-2015
[7] Frege et al., 2017 doi:10.5194/acp-17-2613-2017