Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2021-735-RC2, 2021


# Review of Jokinen et al. (acp-2021-735)

Anonymous Referee #2

---

Referee comment on "Measurement report: Long-term measurements of aerosol precursor concentrations in the Finnish subarctic boreal forest" by Tuija Jokinen et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-735-RC2, 2021

---

This measurement report by Jokinen et al. quantifies the seasonal and monthly variation of the important aerosol precursor gases sulfuric acid (SA), methane sulfonic acid (MSA), iodic acid (IA), and highly-oxygenated organic molecules (HOMs) from the remote field station SMEAR I in Värriö Northeastern Finland. The chemical species are measured almost constantly by a state-of-the-art nitrate chemical ionization atmospheric pressure interface time-of-flight mass spectrometer (CI-APi-TOF-MS) in the period between April-October 2019. In addition, the authors link the four chemical species and their seasonal concentrations, as well as meteorological parameters to new-particle formation (NPF) events classified by a Differential Mobility Particle Sizer (DMPS) and a Condensation Particle Counter (CPC).

The overall measurement reported is of good quality and its presentation is well structured, while the language is for the most part precise and fluent. As the authors mentioned, these are the first long-term measurements in a sub-Arctic region that can help the scientific community to understand regional chemical processes and formation of atmospheric aerosol as well as their dynamics in this remote region that is very sensitive to the Earth's rapid climate change. The scientific methods and assumptions are valid, and the interpretation of the measured data is discussed in detail, while the conclusions are limited and not always entirely clear. The conclusions therefore rather give a rough overview of the detailed mechanism of new-particle formation in sub-Arctic areas. As a measurement report, however, this work provides some interesting insights and serves as the basis for future and expanded research in this region. Since there are no major scientific concerns, I believe that this manuscript can be published in ACP as a measurement report after addressing the following mostly minor comments.

***Minor comments:***

- line159-61: Does this LOD of $4x10^4/cm^3$ also applies to other species than SA?
- line197-99: Does these heatwaves have a substantial impact on the NPF at this site? Can the authors elaborate a little more on this in the discussion part as this may more often in the future?
- Figure 2: Since anthropogenic pollution plumes from the nearby smelters also affect the SMEAR I site, mixing ratios of $SO_2$ and $NO_x$ should be added here, if available, like it is done in Sipilä et al. (2021). This can help to understand various aspects of the photochemistry at this location (e.g., types of radicals and quantitative estimation, such as OH, $NO_3$, $RO_2$) and to separate NPF events from biogenic and anthropogenic sources.
- line239-41: Why does the missing March data make the comparison (*more*) uncertain and in what perspective/direction. Can the authors elaborate on how far the diurnal cycle (e.g., peak concentration) or the overall SA concentration might change?
- line243-53: Is there $NO+NO_2$ mixing ratios available during this period to state whether the HOMs at SMEAR I are mainly non-nitrate HOMs or organonitrates? This difference can also have a strong impact on HOM dimer formation and thus on NPF at this site. Taking the sum of HOMs (non-nitrate and organonitrates) from Sulo et al. (2021) at SMEAR II would result in a comparable diurnal cycle of total HOMs to SMEAR I. Sulo et al. (2021) shows a clear peak of organonitrates in summer at midday around $1x10^7$ mol./$cm^3$ while the non-nitrate HOMs are at ~ $0.4x10^7$ mol./$cm^3$. In the evening non-nitrate HOMs increase to about $0.8x10^7$ mol./$cm^3$ while the organonitrates begin to decrease but remain almost at the same concentration as the non-nitrated HOMs. It would be easier to follow the discussion and improve the reader's understanding of the various chemical species to include this section on HOMs in the later section starting at line 314.
- Figure 3: It might be useful to add the diurnal cycle of the global radiation from the different seasons within the plots to link the aerosol precursor gases production to the ongoing photochemistry at SMEAR I station.
- line314-16: Is the sum of HOM concentration in Yan et al. (2016) similarly calculated to this study (summation between 300-600 Th) and corrected for transmission to be comparable? Please add Yan et al. (2016) to the reference list.
- Figure 5: I would also recommend plotting HOM concentration on the x-axis and temperature on the y-axis while coloring the global radiation. This might also reveal whether the behavior is linear or else. Does one data point represent a daily mean?
- Figure 6+7: Since there is a clear seasonal (and even monthly) variation in aerosol precursor gases concentration, I wonder if this also leads to a diurnal variation of NPF events. Therefore, a more distinct seasonal (or monthly) profile of median number size distribution and total particle concentration of the NPF events here would be more meaningful in my opinion.
- line364-71: The authors may add distinct numbers to the temperature, RH, and global radiation differences between event- and non-event days, as was done for the ozone concentration.
- line384-87: To what extent are the boundary layer dynamics affected during the day; especially concerning sunrise and sunset during the different seasons. What influence does it have on the measured precursor gas concentrations?
- line391: Since the concentration of the measured gases depends on the wind direction and the air masses exposed to sources of volatile precursors, it would be great to add the wind direction to Figure 1.
- line394-401: It looks like the air masses of cluster3 of event days and cluster2 of non-event days passed over the highly polluted region of Kola Peninsula. A representation of the average particle number size distribution as a function of the different air mass clusters could therefore be very helpful here. In addition, as noted by Reviewer 1, the values of the condensation sink and the coagulation sink of small particles are important parameters for the classification of NPF events that should be added to the manuscript.
- Chapter 3.3: It would be helpful to have an overview showing the aerosol size distribution, wind direction, concentration of small particles or 2-7 nm negative ions,

the particle sinks, and the NPF events over the entire measurement period, similar to Figure 2 in the accompanying article by Sipilä et al. (2021). In addition, the NPF events can be highlighted in Figure 1 and Figure 2 using background shading.

- line409-414: Instead of using the sum of the HOMs, it may be useful to focus only on the ULVOC and ELVOC parts when parameterizing NPF, if possible. As mentioned by the authors, however, HOM may play a minor role in nucleation, while it can strongly influence particle growth and compete for coagulation losses.
- line415-417: Is there no evidence of mixed clusters between sulfuric acid, iodic acid, MSA, HOM, or possibly bases such as ammonia or amines observed by the nitrate CI-APi-TOF as reported by Sipilä et al. (2021) with the APi-TOF?
- To maximize the information provided in the abstract, please include unambiguous numbers of aerosol precursor gas concentrations and measured values rather than relative descriptions (e.g., "MSA shows a more distinct seasonal cycle with concentrations peaking in the summer of about $1 \cdot 10^8$ mol./cm$^3$"; or: "under relatively low temperature (1-8 °C)"). Why is the significant higher global radiation on NPF-event days not mentioned in the abstract?

***Technical corrections:***

- Please add the numbering of the individual chapters according to the text.
- Some passages require language editing.
- line170: […] were downloaded […]?
- line247: missing space between 'are one'.
- line298: missing 't' in events.
- line336-337: "[…] and detected aerosol precursor gases during NPF days […]."
- Figure 6: In the figure caption there is a 'during' too much.
- line369: "The meteorological conditions that favor NPF are thus similar to those at the SMEAR II station in Hyytiälä, […]".
- line377: Where does the reference 'Kulmala et al. (2013)' belong to? Furthermore, in this line add a 'C' to figure reference (e.g., Figure 8C).
- line467: SMEAR III?
- The short summary needs to be revised in the second sentence.