REVIEWER 1

General Comments

This paper by Thakur et al. explores new particle formation events in Helsinki from gaseous **precursors of marine (iodic acid, sulfuric acid) and anthropogenic (sulfuric acid) origin. Importantly, this study highlights the complexity of nucleation in a semi-urban location with marine and anthropogenic influence. The authors use a wide range of ground-based instruments to monitor particle size and concentration, in addition to measurements of key gas-phase species. These measurements are paired with meteorological and satellite observations to identify the source of the precursors to NPF. This study fills a measurement gap of nucleation events in coastal urban areas.**

This paper has some interesting results that are valuable to the NPF community. With that, I find it suitable for publication in ACP. However, this paper would benefit from clearer explanations of how the conclusions were reached, or perhaps some softening of their conclusions. Furthermore, I believe this work could use some editing for clarity.

We thank the reviewer for appreciating the work and providing the specific and very valuable comments which has considerably improved the quality and clarity of the manuscript. We have answered the queries/comments for each point as detailed below. The corrections would be incorporated in the revised manuscript including softening of the conclusions reached in this work.

Specific Comments

- **In the abstract (L49), the authors indicate that the type of phytoplankton species and the intensity of the bloom was one of the most important factors affecting aerosol precursor vapor concentrations (IA and SA). How was this conclusion reached, when the only measurements made in this study to link their gas and particle phase measurements to biological activity were satellite measurements of Chl-a,which does not differentiate between species?**

We agree with the reviewer, that we did not make any actual measurements of the algal species neither did we do any species identification for this study. But we speculate that the emissions from the type of phytoplankton species found in a particular area (area selected based on the trajectory of air mass) might be influencing the gas concentrations in the atmosphere. Our interpretations are based on the residence time of air masses in a particular marine region. We made the best possible estimations on

the species present in that region based on Baltic-wide monitoring of cyanobacterial blooms from previous studies mentioned in the MS. As per these studies (Knutson et al., 2016; Attard et al., 2019; Kownacka et al., 2020) results show that bloom composition is fairly consistent for different regions and seasons from year to year, which makes it possible for us to make close estimations of the species present during our study in a particular region (from where the airmass travels). Importantly, however, the bloom composition during summer is different from spring time blooms, which we detail in our study and helps us interpret particle formation and their potential sources.

Accordingly we have added /modified the statements in the section 3.4

"The Gulfs of Bothnia and Riga are dominated by the genus *Aphanizomenon* (Kownacka et al., 2020). In addition, the Bothnian Sea and Gulf of Finland were found to be rich in cyanobacterial genera of *Aphanizomenon* along with *Nodularia* and *Dolichospermum* (Kownacka et al., 2020). As per the previous studies which carried out the Baltic-wide monitoring (Kowancka et al., 2020 and the references mentioned therein) that bloom composition is fairly consistent for different regions and seasons from year to year, which makes it possible for us to make close estimations of the species present during our study in a particular region (from where the airmass travels and the residence time over a particular region).

- **The authors also conclude on L696 that the type of phytoplankton species, bloom intensity, and distance from the bloom plays an important role.**

    1. **How does the phytoplankton species affect the gas-phase concentration in their measurements?**

       Not all plankton species emit DMS (a precursor for biogenic SA). There are only very few specific species found in some particular areas that may be relevant to account for regarding their contribution to biogenic SA in the atmosphere. Similarly, specific species of macroalgae are responsible for large emissions of I_2 which finally oxidizes to IA either at the source or during their transport to the study site. A detailed explanation of the species and their niche is explained in section 3.2.

    2. **Is there an instance where there was a sea wind with less intense phytoplankton bloom, and no NPF events?**

       Yes, we observed a few days when this occurred; for example, on August 13 and August 17 when there was no event, yet the there was a sea wind and the bloom was less intense as

compared to other event days. An example of such a day (August 17) is shown in the figure S13. The figure and this required explanation is incorporated in the supplementary information as reference.

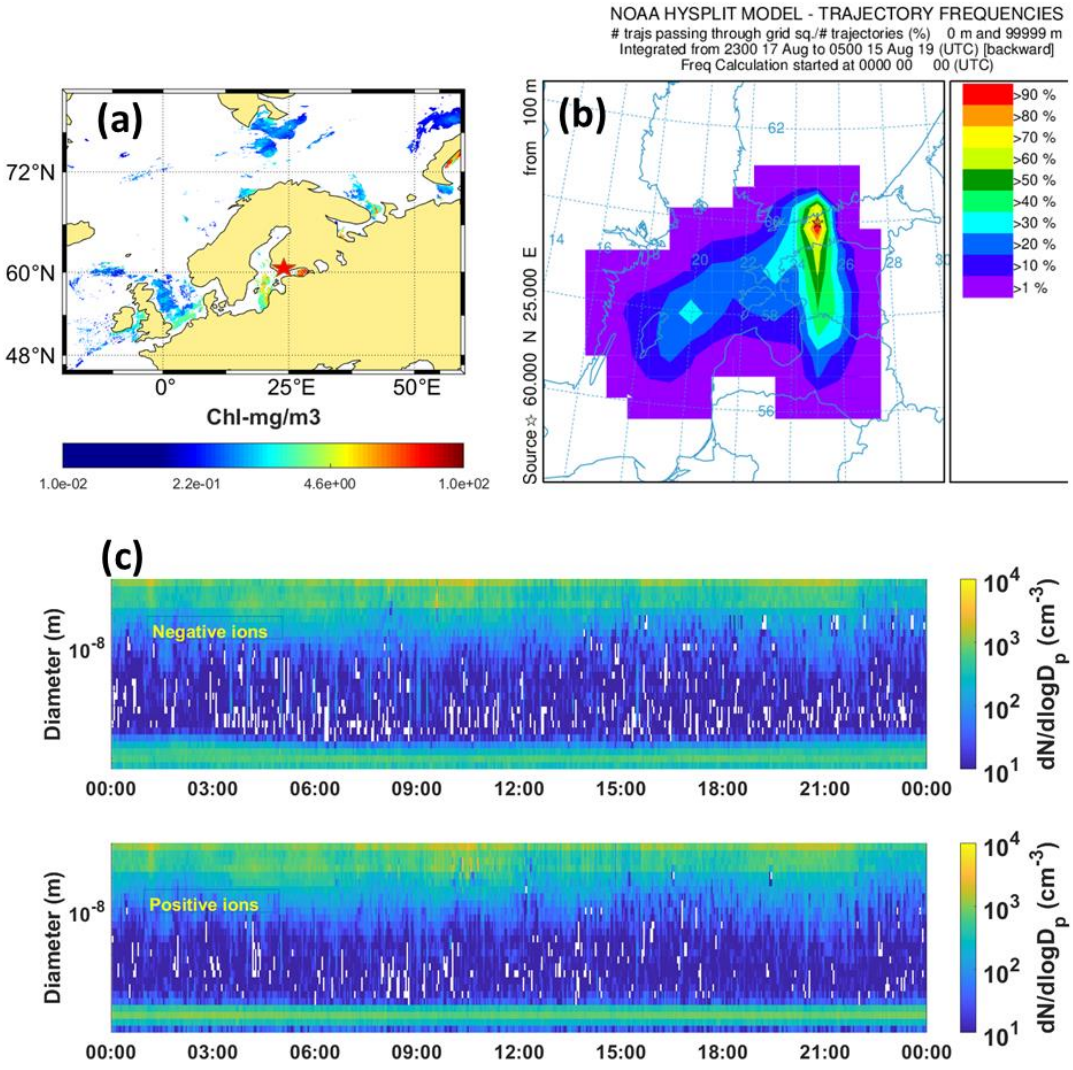

Figure S13: No event day on 17 August 2019 (a) Chl-*a* concentrations (MODIS), the red star shows the experimental site (b) Trajectory frequency plot (100 a.g.l) for 24 h back trajectory using GDAS meterological input data (frequency grid resolution: 1.0° × 1.0°) (c) Charged particles number size distribution (negative: upper, positive: lower) obtained from the NAIS.

- **I'm also not convinced by the importance of the cyanobacterial blooms on the IA concentration, especially when compared to the other algae and marine sources. The authors timed their study to match with the cyanobacterial blooms that are expected in the Baltic Sea and coastal regions of Finland. In section 3.1.2 however, the authors emphasized that the cyanobacterial blooms were reduced below normal in July and August, which were the time periods in which they**

observed the NPF events. The authors also point out that the low tide and high irradiance could be a source of macroalgae iodine, as was observed in McFiggans et al., 2010.

- In section 3.3.2 (Case 1) we propose that the contribution of macroalgae to IA could be the dominant source when IA is speculated to play a dominant role in initiating the burst event on 11 August. However, we have based our conclusions only on the high Chl-*a* values in the region from where the air masses originated and where the air mass residence time was the highest. This section does not talk about the dominant contribution of IA from the Cyanobacterial blooms.
- In section 3.3.2 (Case 2): However, we speculate that when the wind direction (coinciding with high residence times) was over the bloom areas which are dominated by specific cyanobacteria producing DMS (a precursor to SA), we see an increase in SA followed by a burst event. The interpretation regarding particular species of cyanobacteria in the respective marine and coastal areas is based on ongoing yearly cyanobacteria monitoring coordinated by the Finnish Environment Institute (and cited in the text). No species identification was done for this study. However as stated above the bloom composition is relatively stable for particular seasons and areas between years, but their intensity may vary depending upon temperature and nutrient availability.
- Section 3.1.2: It is correct that cyanobacterial/algal blooms were less intense at end of July and August. And it is worth noting that when the blooms of both cyanobacteria and indeed macroalgae start to decay and die (while being exposed to sunlight) they produce more emissions (biogenic SA and IA). Thus, this itself is a reason for speculating why we see most of the NPF events in later summer months. All these observations suggest that there could be a strong link between algal/cyanobacterial emissions and their impact on NPF. However, as stated in our conclusions further studies are definitely needed to confirm these findings in a coastal setting.

  In order to clarify this point we have included the following lines in the conclusion section of the study:

  “In fact, an overall higher impact of biogenic emissions was noted in this semi-urban site particularly during end of July and mid-August when the bloom intensity decreases and the cyanobacteria/macroalage start to decay and die, while being exposed to sunlight, they produce more biogenic emissions biogenic of SA and IA”.

Would Chl-a measurements also measure the contribution from macroalgae?

Yes the satellite Chl-a measurements can indicate if the contribution is from Macroalgae in case of high values (higher than the average) of Chl-a. This might indicate that there are floating/exposed macroalgae present. However, typically these algae are not present in the open sea areas of the Baltic Sea and the resolution of the satellite Chl-a are not in such high resolution as to permit an interpretation (or differentiation) with higher confidence, hence it's not mentioned in the MS. Nevertheless, the Baltic Sea has a great abundance of macroalgae along its coasts that we speculate could be contributing to the IA signal.

- **L485: The authors indicate that the change in wind direction 'apparently discontinued the precursor vapor source', however I'm not sure why this is apparent? From Figure 5(d), the concentrations of SA, MSA and IA remain relatively constant with the change in wind direction.**

We thank the reviewer for pointing this misinterpretation in the manuscript. We accept that the changed air-mass just discontinued the growth and not the precursor vapor concentrations. We have clarified this and changed the lines to

"This shows the particles must be the process of growth mostly elsewhere, which is not evident in the changed air mass, however we still observe almost the same (or even slightly higher) precursor vapor concentrations, since the wind still passed over the bloom areas before entering our study site".

- **Figure 6: Is the green trace called 'particles' the measured particles? Perhaps make that more clear.**

The figure caption states that these are observed particles (measured through NAIS). However, we have changed the Legend to "measured particles (NAIS)" to make it clearer.

- **L593: How do you know all the I2 was oxidized to IA?**

The reviewer has correctly pointed out that not all I_2 is oxidized to IA. Also, in the present study we cannot give an estimate that how much I_2 (from source region) would be converted to IA. Hence the lines 660-661 have been changed to

"By the time the air mass reached our measurement site from the emission source, a fraction of the emitted I_2 could have oxidized to IA".

- **The authors often use parentheses to provide additional details within the text. In some cases, the parentheses are unnecessary and interrupt the flow of the text. I suggest the authors review their use of parentheses for clarity. Some examples:**

    1. **L32: Several studies have investigated New Particle Formation (NPF) events from various sites ranging from pristine locations, including (boreal) forest sites to urban areas.There have been studies of more than just boreal forests, I'm not sure why boreal was specified here. Can remove the parentheses and/or the word boreal.:**

       The parentheses has been removed as per the reviewer's suggestion.

    2. **L101: The parentheses around 'produced from macroalgae' can be removed**.: The parentheses has been removed as per the reviewer's suggestion.
    3. **L499: Can be rewritten as 'The high normalized signals…' to remove the parentheses.:** The parentheses has been removed as per the reviewer's suggestion.
    4. **L355: Can use 'The daily mean' instead of The mean (whole day):** We corrected the statement throughout the MS as per the reviewer's suggestion.

Technical Corrections: *All the technical corrections have been incorporated in the revised MS*

L42: Keep the chemical names in lowercase "sulfuric acid (SA)" to match L78

- It has been corrected in the MS as suggested by the reviewer.

L46: Chemical names in lowercase "iodic acid (IA)"

- It has been corrected in the MS as suggested by the reviewer.

L150: Use the abbreviation for New Particle Formation (NPF)

- It has been corrected in MS as suggested by the reviewer.

L196: I'm not sure what 'mlpm' is, define it?

- mlpm stands for milliliter per minutes, we have corrected these unit to "mLpm" in the revised MS

L205: Define HOMs when it is first used

"Please note that the concentration of highly oxygenated molecules (HOM, monomers and dimers) were calculated from the unit mass resolution data".

L208: extra 'The'. Don't need to define UMR if you only use it once.

We have made the correction in the MS as mentioned in the sentence above.

L263: Don't need to redefine growth rate.

- It has been corrected in MS as suggested by the reviewer.

Table S1: O_3 instead of O_2?

Yes we mean O_3, thanks for pointing out this typo error. We have corrected it now to O_3.

447: Missing a period?

Period added now in the revised MS. Thanks for pointing the error.

556: Replace HIO_3 with IA

We replaced HIO3 with "IA".

Figure 4 caption: Not sure what the yellow circles are for 'all time' – is it just the other time except the morning and evening?

Yes that is correct, that yellow circles denote "all time". It is mentioned in the Figure caption also, as underlined below.

"Figure 4: Correlation of SA with MSA (a,b), SO_2 (c,d) and NO_x (e) for June–July. The black dashed lines for both axis represent the mean of the gas concentration, red dashed line represent the median value the gas concentrations and red solid line represents the linear fit. Spearmann's coefficient (r_s) was used to test the correlation, at significance level, 0.001. The circles represent data points at different hours of the day. The upward pointing green triangles represent the morning rush hours (6:00–8:00 h) and the downward pointing blue triangles represent the evening rush hours (15:00–17:00 h). The yellow hollow circles represent all data. NO_x data unavailable of August"