Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2021-729-RC2, 2021

Comment on acp-2021-729

Anonymous Referee #2

Referee comment on "An evaluation of new particle formation events in Helsinki during a Baltic Sea cyanobacterial summer bloom" by Roseline C. Thakur et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-729-RC2, 2021

Thakur et al. present field measurements of new particle formation events occurring in Helsinki. This site is impacted by air masses from the city and sea. Observations from Helsinki help fill in a critical gap in understanding how marine new particle formation impacts urban air quality. Their observations relate nearby algal and cyanobacteria blooms to marine new particle formation events.

Overall, the information presented in the paper is logical but some of the conclusions on which precursors contributed to which nucleation events are not persuasive. The paper is not written as concisely as it could be with many parts repeated and it's difficult to follow. Only some of these instances have been pointed out here. The authors should try to shorten the paper. The study fits well in ACP. Several aspects of the manuscript should be improved prior to acceptance for publication.

Major Comments:

Line 124: Why are more coastal measurements needed? The authors detail out a few studies conducted at the coasts and where they found correlations to coastal seaweed and algal blooms. What does this study add to the scientific field other than more measurements? How do measurements from Helsinki help the scientific field? I am sure these measurements are important but framing the "why" will help the reader better understand the purpose of this study.

Line 199: The authors state that the CIAPiToF was calibrated following the procedure detailed in (Kürten et al., 2012). That study only calibrated the CI-APi-ToF for sulfuric acid. How does this calibration constant apply to MSA, iodic acid, and organics with nitrate

as the chemical ionization reagent ion? What is the systematic uncertainty associated with using this calibration constant for non-sulfuric acid molecules? Often the authors report 3 significant figures on their precursor concentrations. Is this in-line with their estimated uncertainty?

Along these same lines, what was the holdover time of SA, MSA, and IA (and other compounds) in the CIAPiToF inlet? On line 599, the event lasted less than 30 minutes. These compounds are very sticky and likely persist in the sampling lines even if the sampling rate is high. They likely persist at different rates so the order at which each compounds reaches its maximum concentration (and its absolute concentration at the maximum) will vary. Have the authors examined this to better determine if short burst new particle formation events can actually be studied with this instrument setup? How would time dependent wall loss rate impact the calculation of growth rates?

Line 322: how long did the cyanobacteria bloom last during the measurement campaign? What area did it cover? In line 324, what does lower than normal mean? Lower than June? Some numbers would help put this intensity in perspective. In line 582, the authors comment that the blooms are intense but how does this compare to other periods of time. Is there a correlation of bloom intensity with IA, MSA, and SA concentrations (assuming the air mass is coming from the bloom's direction)?

Line 331 and paragraph beginning 633: Did the authors actually measure the algae and cyanobacteria types during the blooms that occurred during the measurement period? Bloom composition can easily change based on numerous factors so it may not be a fair conclusion to link these previously measurements bacteria and algae types to what was observed during the measurement campaign. How confident are the authors that they can link these algal species to their new particle formation events?

Line 471: From previous NPF campaigns, it seems that sulfuric acid concentration should increase before observed particle concentration? The text suggests the sulfuric acid concentration increased after particle concentration increased. Figure 5 shows that SA was already increasing. Did the authors observed any freshly formed clusters with the CIAPITOF? From line 499, it seems the authors did observe some clusters (and shown in Figure S4). It would be helpful/more logical to mention this earlier. Did the authors measure DMA and ammonia concentrations? If the authors believe SA-DMA is forming neutral clusters, do the authors know where the DMA is originating from? Why does the SA+DMA cluster concentration peak significantly after the new particle formation event (figure S7)? On line 474, what is a local clustering event? Does this refer to clusters observed on the CIAPITOF?

Line 520: Why does MSA and IA concentrations need to increase in order to demonstrate they could participate in that NPF event? Also, from Figure S5, the concentrations of both are increasing (before the signal cuts off). How do the authors know for certain that these compounds are not participating in the event? Also HOM concentrations do not seem any higher than those in Figure 5. So why is this event SA-HOMs driven? What clusters did the CIAPITOF see? The organic clusters shown in Figure S4 just show a constant and slow

increase in organic cluster concentrations throughout the day.

Line 558: Isn't the temperature during this campaign much higher than 10C? Higher temperature will still favor SA+amine/ammonia nucleation. Observations of HNO3.IO3- and H2O.IO3- clusters doesn't indicate IA nucleated. Were larger IA clusters seen? Also did the authors calculate how much IA contributes to growth? Is that why the authors are implicitly linking high concentrations of IA to particle growth in line 564?

Line 567 (And figures 5,7,8): The authors comment that the Aitken mode particle concentration increase after a new particle formation event. Why does the concentration drop before+during a new particle formation event? No doubt the decrease in scavenging rates allows nucleation to occur but what is leading to this drop in large particle concentration? This seems just as important as an increase in precursor concentration in producing an event.

Minor comments:

Line 154-160: It would be less confusing if the instrument details in this paragraph moved to the instrument 2.1. Otherwise the reader will want more details about the instruments before the actual instrument section.

Line 170: the paper hypothesis has already been stated in introduction. No need to state it again here.

Paragraph beginning 169: It would be more useful if this paragraph focuses instead on presenting the date+times of the algae and cyanobacteria blooms during the measurement campaign. The background on why there are more blooms should be mentioned in the introduction instead to better motivate this study.

Line 189: What type of inlet?

Line 195: Was the only reagent ion NO3-? Or did it have ligands?

Line 196: is mlpm milliters per min? The L should be capitalized to make it less confusing. Or ccm which is more commonly used? Or maybe mlpm is fine? But it was initially confusing to me.

Line 208: There is a random The at the end of the line

Line 213: What do the authors mean by two identical DMAs? Have they quantified the transfer functions and transmission efficiencies for both to say they are identical?

Line 280: Are these mobilities diameters?

Section 2.5: Condensation sink spans what particle diameters? (I think it's >6 nm). Is there a reason why CS does not include surface area of smaller particles which could be significant during a new particle formation event?

Line 340: is open sea microalgae cyanobacteria? Can the authors more clearly show/explain what time periods were for coastal macroalgae and blue green algae? Did they these bloom/exposure events overlap? If so, to what extent?

Line 406: For surface emissions to be carried to the measurement site, the surface wind speed is important. Is this wind velocity at the surface/altitude of the measurement site? Or does it include a component of vertical velocity? In other words, how confident are the authors that the air mass is not vertically mixing downwards which would dilute the surface emissions?

Line 424: MSA also originates from agriculture.

Line 459: could a burst in sub-3 nm particles be from a suppression in growth and not a local nucleation event?

Figure comments:

Figure 2: Is this figure necessary? The manuscript only details specific events that occurred in short periods of time. It would be more helpful to see this data with the event data.

Figure 5,7,8 (and their siblings in the SI) are very difficult to read. The font on the labels is too small to read. It might be easier to have the timeline graphs vertically stacked so it's easier to compare between them. The F panel is strange. Are the maps of the same area? It doesn't seem like it. Why have two panels for F?

Figure S3: The labels are too small to read. Units of residence time?