Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-390-RC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric new particle formation characteristics in the Arctic as measured at Mount Zeppelin, Svalbard, from 2016 to 2018" *by* Haebum Lee et al.

Anonymous Referee #2

Received and published: 17 June 2020

"Atmospheric new particle formation characteristics in the Arctic as measured at Mount Zeppelin, Svalbard, from 2016 to 2018" by Haebum Lee et al.

General:

To assess the effect of ocean biology on atmospheric new particles formation observed between 2016-2018 at the Mount Zeppelin, Svalbard, this study was set to explore (using MODIS monthly mean satellite Chlorophyll-a concentrations) whether any direct link or correlation exist between ocean biological sources and the observed frequency of occurrence of nanoparticles as small as 3 nm diameter.

It was concluded that nanoparticles increased more frequently when the origin of

air masses reaching the Zeppelin observatory overlapped with regions having strong Chlorophyll-a concentration and dimethyl sulfide (DMS) production capacity, and was also correlated with increased daily NH3 concentrations. Moreover, the authors argue that the primary drivers of the observed new particles formation are the seasonal cycles of ocean biological activity or presence of sea bird colonies. However, it's essential to keep in mind that overlap or correlations do not prove a causal relationship. Unfortunately, the authors do not present any assessment of the reliability and credibility of the derived correlation.

Moreover, the literature survey is not fully convincing; there is some previous observational evidence on new particle formation from the Zeppelin observatory that reflect the seasonal cycle of gel-forming marine microorganisms and their controlling factors, that would seem to have an essential bearing on the results obtained and, thus, appear to merit discussion. Examples are Heintzenberg et al., 2017; Karl et al., 2019 and Mashayekhy Rad et al., 2019.

Furthermore, the satellite derived Chlorophyll-a numbers used in this study cannot be used in order to demonstrate a biological source in terms of biological activity (productivity/bloom) or phytoplankton primary productivity for the following reason; Chlorophyll-a is used a proxy of phytoplankton biomass whereas a phytoplankton bloom in various phases is the combined effect of phytoplankton production and zooplankton grazing, cell lysis and/or bacterial degradation. To be able to make any kind of statement relating the biological activity, phytoplankton biomass- and speciation, zooplankton etc. to DMS water concentrations, data on these parameters are required, covering the period 2016-2018.

The generally, over marine areas, poor correlation observed between parallel measurements of seawater DMS and phytoplankton biomass (Chlorophyll-a) has been explained as a consequence of the species specificity of DMS production followed upon the production of from extra-cellular (dimethyl sulfonium propionate) DMSPp.

A further very important reason for the in general poor correlation between DMS in the water and in algae biomass is that DMS is produced from DMSPd and not from intra-cellular DMSPp. This means that the production and breakdown of DMS in the water column must be looked on as a result of complex physiological and ecological interactions, as demonstrated in Leck et al. (1990). To be able to make any kind of statement linking the biological productivity, phytoplankton biomass and DMS water concentrations, data on the above-mentioned parameters are all required. As no such measurements were performed it is not valid to extrapolate results of high satellite derived Chlorophyll-a concentrations in the surface waters to DMS in the water column which will be subsequent emitted into the atmosphere and there undergo photochemical oxidation to form various intermediate products and, ultimately, sulphuric acid promoted nucleation.

Therefore, the statements causally linking the effect of ocean biology or as referred to in this study "DMS production capacity" on atmospheric new particles formation observed at the Zeppelin observatory are not well founded or supported and should be removed or de-emphasized.

As the conclusions are based on essential inputs of MODIS monthly mean satellite Chlorophyll-a concentrations and the suggested presence of bird colonies, which are both not constrained by measured in-situ data and that it seems that the authors have a somewhat limited conceptional understanding of the detailed processes involved needed for a successful assessment the effect of ocean biology on new particle formation observed at the Zeppelin observatory, I could only recommend the manuscript for publication after major revision according to the given comments and suggestions.

Although many of the above-identified parameters are not given as an integrated part of the present study, the manuscript has an important value as a descriptive data report of the seasonal life cycle of nanometer sized particles down to 3 nm diameter (with a relatively high temporal resolution), a parameter in general sparsely measured in the marine environment and specifically so in the Arctic.

Detailed:

Page 2, line 50: relevant reference to add is Karl et al., 2013. Page 2, line 63: Please omit Covet et al., 1996. Page 3, line 68: Please add Heintzenberg et al., 2017. Page 3, line 72: Please insert number between measured and size. To not confuse the reader be specific on that you discuss size distributions by number, throughout the manuscript. Page 3, line 78: Please remove the superscript (10) on "occur".

Page 3, line 80: Please motivate why you will use satellite-derived Chlorophyll-a concentration data to detect potential source regions for new particle formation. Please also add that Chlorophyll-a is a proxy of phytoplankton biomass only.

Page 3, lines 90-93: Observations are a challenge and specifically so in the pristine remote Arctic marine environment. Specific to the Zeppelin observatory extreme care must be exercised to prevent interference from local pollution (ship traffic) and thus contamination of the air samples. Please add information on the implemented procedure to detect and avoid contamination by local pollution or from long-range transport (southerly air mass origin). Did you have any automatically interruption of the sampling when necessary, due to unfavorable conditions (pollution sensor)?

Page 4, line 97: General to the manuscript. Please replace specie(s) with compounds or constituents as specie(s) belongs to the family of living organisms.

Page 4, line 97: All Ionic – and molecular formulas should be define.

Page 4, lines 97-101: Please give the 50% cut-off equivalent aerodynamic diameters (EADs) of the 3-stage filterpack sampler (type?). Also give details on the analytical methods used for both the particular matter and gas phase compounds collected. How were the blank levels determined? Analytical detection limits obtained for the various ions? Were any Quality checks of the IC-analyses performed? LOD, precision?

Page 4, line 102: Please define AWIPEV.

Page 4, line 106: Please define EMEP, ACTRIS, GAW-WDCA.

Page 5, line 142: Please add details on how the sulfuric acid number concentration was predicted from the measured daily SO2, please also discuss the quality of the data in use.

Page 5, line 154: Be specific, which are the precursor gases in mind? How do you verify their abundance? How will a proxy measure of marine phytoplankton biomass (Chlorophyll-a) influence the availability of atmospheric precursor gases for new particle formation?

Page 6, line 176: Please add the results by Heintzenberg et al., 2017.

Page 6, line 178: Please clarify which results that support the following statement “In addition, DMS originating from marine sources can be a key precursor contributing to NPF in the remote marine atmosphere”.

Page 6, lines 181-182; Page 7, lines 203-204 : How do you explain the poor correlation observed between the highest percentage of new particle formation (Fig. 5), and the relatively low MODIS monthly mean satellite-derived phytoplankton biomass (Chlorophyll-a) concentrations in the month of August (Fig. S1 bottom and Fig.6)?

Page 6, line 187: I cite “In addition, the melting of sea ice in summer can increase the availability of marine biogenic sources, promoting NPF”. Please specify which the sources you have in mind? Here I find the literature survey unconvincing; there is quite a large amount of previous observational evidence that would seem to have an essential bearing on the results obtained and, thus, appear to merit discussion. In this respect, it seems somewhat surprising that even not mention of or learn from the previous work by Leck and her colleagues over the last three decades on releases of atmospheric sulfur compounds and marine sea-spray aerosols (organic polymer gels/inorganic) over the Arctic pack ice area (incl. the marginal ice zone) in summer.

Page 7, lines 204-207; Page 8 lines 239-242; Page 9, line 281: Figure 6 shows that the Chlorophyll-α concentrations were most pronounced in the ocean areas southwest and

southeast of Svalbard. After the authors explored the potential source regions of the air masses in relation to occurrence of nanoparticles, it was found that increasing numbers of the latter occurred more frequently when the air trajectories passed over the oceanic regions to southwest and south of Svalbard but surprisingly not when passing over the ocean areas south east of Svalbard. This was explained by that the DMS production capacity of the southwest ocean was 3 times greater than that of the southeast ocean. To support the findings the authors used the results derived from a study based on data sets obtained between May and April in 2010, 2014 and 2015 by Park et al. (2018). Please give a detailed explanation to what “DMS production capacity” stands for and what it critically depends on. Please also explain how the findings by Park et al., 2018 covering only two months (April and May) of the biological season (this study April to October) and in different years, could be used to explain the findings in this study.

Page 7, line 208: Please support your statement: “The existence of significant amounts of nanoparticles as small as 3 nm during NPF events at the study site suggests that NPF occurred there, rather than the particles being transported from other regions after growth.” What is the expected atmospheric residence time of the nucleated particles?

Page 8, line 252: Please clarify the meaning of that non-sea-salt sulfate could have a secondary origin from oceanic DMS. Which formula was used in the calculations of non-sea-salt sulfate. How do you estimate the contribution from non-biogenic DMS sources? A much more strait forward comparison would be to use particulate methane sulfonate (MSA) concentrations of the total suspended particle samples.

Page 8, line 252: Could you please discuss how realistic your assumption on a DMS derived (sulfuric acid) nucleation mechanism is in respect to the findings by Pirjola et al. (2000), which showed that, under typical conditions in the MBL, homogeneous binary H2SO4-H2O nucleation will not occur and ternary H2SO4-H2O-NH3 nucleation will only be sufficiently effective to produce observable particles for DMS concentrations in the range of 400 ppt(v) or higher and very low aerosol condensation sinks.

Page 18, Figure 2: Please remove the superscript (37) on “SRAD”.

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