Comment on egusphere-2022-1079
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Community comment on "Late summer transition from a free-tropospheric to boundary layer source of Aitken mode aerosol in the high Arctic" by Ruth Price et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-1079-CC2, 2022

General:

Although, the manuscript cites many very relevant articles from the field the literature survey is not fully complete; there is quite a large amount of previous observational evidence that would seem to have an essential bearing on the results and conclusions obtained and, thus, appear to merit discussion. In this respect, it seems beneficial to mention of or learn from the previous work by Tjernström and Leck and their colleagues over the last three decades over the summer Arctic pack ice area (incl. the marginal ice zone).

Their work has shown that it is not only necessary to be able to specify particle concentrations in the atmosphere and their possible sources, also we must understand the thermodynamic structure of the lower atmosphere (typically a well-mixed shallow boundary layer at the surface, only a couple hundred meters deep, capped by a temperature inversion below the free troposphere), the dynamics of the boundary layer, and processes important in exchange between the air and ocean top layers.

It should be made clear that this contrasts with the processes in more southerly latitudes, where deep convection, could enhance mixing across the whole troposphere. Over the pack ice this does not occur other than possibly in frontal zones associated with passing weather systems.

As such, the structure of the pack ice lower atmosphere generally limits mixing of aerosol particles in the free troposphere into the boundary layer. For this to happen it requires that aerosols sources in the free troposphere (either sourced locally or from long distant advection from lower latitudes) are brought down to the top of the boundary layer where entrainment can occur, and the only mechanism that can bring elevated plumes down to the inversion is large-scale subsidence, which is a very slow process. Entrainment is thus unlikely to be a major factor contributing to CCN (Aitken/accumulation modes) number concentrations within the lower atmosphere, and thus only will not be a main contributor to the formation of low-level clouds (e.g., Bigg et al., 2001; Tjernström et al., 2012),

These past findings are in direct contrast to one of the main conclusions made in the present study; That particles formed outside the Arctic are the dominant source of Aitken
mode particles during the sea ice melt period up to the end of August, “Particles from such remote sources, entrained into the boundary layer from the free troposphere, account for nucleation and Aitken mode particle concentrations”.

Also not accounted for is the possibility that aerosols and their precursors, lofted in the deeper atmospheric upstream well mixed boundary layer over the open water, could be advected in over the pack ice on top of the shallow local boundary layer (Tjernström et al., 2012) and later be entrained into the local boundary layer through the cloud top by cloud induced mixing (Igel et al., 2017). Other past observations over the pack ice (during melt and beginning of freeze-up) that should have merit discussion is the demonstration that organics found in Aitken and accumulation mode aerosols and in cloud water behaved like marine polymer gels originating from the surface microlayer on leads (open water between ice floes), due to the activity of ice microalgae, phytoplankton and perhaps, bacteria (e.g., Leck and Bigg, 2005; Bigg and Leck, 2008; Orellana et al, 2011; Hamacher-Barth et al., 2016).

Another essential bearing on the conclusions obtained not mentioned is that the thermodynamic structure of the pack ice lower atmosphere has been characterized by two predominantly near-neutrally stratified layers below the main capping inversion of the boundary layer; one in the lowest few hundred meters and one around one kilometer or slightly higher and the possibility of a recoupling and turbulent mixing between them. The reason for the two-layer boundary layer structure is likely a combination of surface-based turbulent mixing from below and cloud-top buoyancy-driven mixing from aloft (e.g., Shupe et al., 2013; Brooks et al. 2017).


Leck, C., and E.K. Bigg, 2005a, Biogenic particles in the surface microlayer and overlaying atmosphere in the central Arctic Ocean during summer, Tellus 57B, 305-316.


**Detailed:**

**Line 20-21:** "Clouds are a major control on the surface energy balance in the Arctic. Due to the low solar insolation and high albedo of sea ice in the high Arctic...” This is true but
only valid over the pack ice area ca north of 80°N. Please clarify.

**Line 31:** Define “high Arctic”. If north of 80°, describe the sources of anthropogenic primary emissions.

**Line 34:** Clarify what is meant by “thermodynamically easier”.

**Lin 36-39:** Make clear that the references to back up the statement concerning the aerosol seasonal cycles are only valid for latitudes south of the Arctic pack ice.

Relevant papers to be added are:


**Line 46:** Please add Heintzenberg et al., 2017

**Line 95:** It should be made clear to the reader that these observations are made over the high Arctic pack ice area. The break-up theory was first introduced by Leck and Bigg, 1999 followed up by Leck and Bigg, 2010, please add prior to Lawler et al., 2021.


**Line 110:** Bulatovic et al., 2021’s modeling study was set out to to explore if Aitken mode particles can act as CCN and influence the cloud properties when accumulation mode aerosols are low in number. The aerosol particle concentrations used in the simulations were chosen to cover a range of typical aerosol size distributions encountered in the summertime central Arctic during four previous campaigns in summers of 1991, 1996, 2001 and 2008 (Heintzenberg and Leck, 2012).

Based on simulated median supersaturations between 0.2 and 0.4 % ranging up to 1 %, the calculated activation diameters were as low as ~ 30 nm, suggesting that Aitken mode aerosols could be activated. Further, the authors examined the representativeness (i.e. how frequently these types of distributions occur in the observations) of the assumed conditions with low accumulation mode concentrations (i.e. lower than 20 cm\(^{-3}\)). For two classes of Aitken mode number concentrations 100 < AIT < 200 cm\(^{-3}\) and AIT < 25 cm\(^{-3}\), the occurrence probability showed to be of 5% and 17% of total minutes of observations, respectively. As such there seems to be a low probability for the combination of low numbers of accumulation mode particles and high numbers of the Aitken mode particle concentration.

Please add the reported frequency of occurrence of observations supporting the simulated activation of Aitken mode particles.


**Line 110:** Please replace Vüllers et al., 2020 with Leck et al., 2020.

Line 118: “the long time series” relative to what?

Line 120: Heintzenberg et al., 2015 gives a detailed discussion on possible aerosol sources for the central Arctic pack ice area based on observations from previous expeditions in 1991, 1996, 2001 and 2008. Please add their reported results to the discussion.


Line 228-235, Equation (3): In the steady-state model of Baccarini et al. (2020a), nucleation of iodic acid is missing as sink. At a nucleation rate of $1/cm^3/s$ and two $\text{HIO}_3$ molecules in the activated cluster it is around 500 molecules $\text{HIO}_3$ per cubic centimeter and second. This might be small compared to the more important losses by dry deposition and condensation but could be relevant if nucleation is strong while number of pre-existing particles is low. At least it should be mentioned that loss by nucleation is neglected in Equation (3).

The uptake of gaseous $\text{HIO}_3$ to fog/cloud water is also not considered in Equation (3).

Line 249-251: It is certainly not clear why the assumption of instantaneous homogenous distribution of $\text{HIO}_3$ throughout the boundary layer had to be made in a global 3-D model. It is very likely that $\text{HIO}_3$ is concentrated at the surface, also because its lifetime is probably less than 1 hour as it mainly depends on the condensation sink typically being in the range of $10^{-4}$ to $10^{-21}/s$.

Line 272: In the XXX_SecOrg case runs, the oxidation rate of monoterpenes was reduced by 100. This is also an unjustified assumption since the oxidation rate of monoterpenes is quite accurately known, maybe with an uncertainty of 10-30 %. Would it not be more likely that gaseous semi-volatile oxidation products that form in the free troposphere are entrained into the boundary layer, molecular diffusion is much more feasible than the entrainment and downward transport of particles.

Line 139: Please add Leck et al., 2020.

Line 160: After concentrations, please add Leck et al., 2022.


Line 274-275: I agree that more work is needed to assess the role of fog in controlling the frequency of iodic acid new particle formation events over the central Arctic pack ice area. However, the control of iodic acid by fog does still not explain the, during past expedition to the same area and at the time of early freeze-up (e.g., Leck and Bigg, 1999; Karl et al., 2013), simultaneous increases in particle numbers occurring in certain size ranges below 50 nm diameter. Also present were accumulation mode particles marine in origin. Stable air masses with at least 4 days or longer residence time over the ice, a surface mixed layer of 100-200m, capped by a temperature inversion and a cloud free stable layer about 1km in depth excluded a tropospheric source.

Line 580: Please replace “The data used here from the ASCOS campaign is available at
www.ascos.se." with “The data used here from the ASCOS campaign is available on the Bolin Centre Database. https://doi.org/10.17043/oden-ascos-2008-aerosol-stratification-1”

**Line 640:** ALL publications relating to ASCOS and AO2018 (MOCCHA, ACAS, ICE) must include the following (minimum) acknowledgment:

“This work is part of the ASCOS and Arctic Ocean (AO) 2018 expeditions. The Swedish Polar Research Secretariat (SPRS) provided access to the icebreaker *Oden* and logistical support. We are grateful to the Chief Scientists Caroline Leck, Patricia Matrai and Michael Tjernström for planning and coordination of ASCOS and AO2018, to the SPRS logistical staff and to I/B Oden’s Captain Mattias Peterson and his crew”.