The study titled "The importance of Aitken mode aerosol particles for cloud sustenance in the summertime high Arctic: A simulation study supported by observational data" by Bulatovic et al. illustrates the impact of Aitken mode particles on summertime Arctic mixed-phase stratocumulus using a series of simulations by two different LES (RAMS and MIMICA). The authors show that Aitken mode particles significantly impact cloud microphysical particles and can contribute to cloud maintenance when accumulation mode particle concentrations are low. The reported results agree with observations from previous summertime campaigns in the Arctic and thus represent a realistic scenario for the high Arctic environment.

The manuscript is generally well written and contains an interesting combination of modeling and observational data. The study adds to our current understanding of aerosol-cloud interactions in Arctic mixed-phase clouds and highlights the importance of small-scale particles for mixed-phase cloud maintenance in the Arctic, which is relatively novel in this regard. Thus, the study has some implications regarding future model studies addressing the cloud response to aerosols in the summertime high Arctic. However, I have a few points that should be addressed before the manuscript is accepted for publication in ACP.

General comments

1. I am missing a section putting the findings of the study into perspective regarding previous work. A lot of studies have been published on aerosol-cloud interactions in the Arctic and the importance of CCN on cloud maintenance has been pointed out previously, especially for the ASCOS campaign (e.g., Loewe et al., 2017, Stevens et al., 2018), which is also simulated here. However, in previous work it was not distinguished between accumulation and Aitken mode aerosols, which is novel to the study presented here and should be pointed out more clearly.

Thus, it should also be emphasized more, <u>when and where Aitken mode particles matter – here</u>, a summertime mixed-phase cloud over pack ice is considered. Is the inclusion of Aitken mode particles only important in those clouds or also for clouds over the open ocean? What about other seasons? From the results presented here it seems like the results are exclusive to summer and pack ice, as in other seasons either accumulation mode aerosols are too numerous (e.g. spring), cloud ice is too high (e.g. winter), and over the ocean Aitken mode aerosols are less abundant; but if this is indeed the case it should be clearly highlighted and discussed.

2. Related to point 1: I would like to see the implications of including Aitken mode aerosol-cloud interactions on the local Arctic environment. As in summer, low-level clouds have an overall cooling effect on the surface which is important in terms of the ongoing Arctic warming, it would be interesting to see the implications on the energy balance at the surface, especially since Arctic Amplification and cloud-radiation interactions seem to be a motivation of this study as mentioned in the Introduction. Also, it would be interesting to see how surface precipitation changes, which could provide information on cloud maintenance beyond the simulated 12 h.

3. To me it is not clear why the model setup differs between the two models (which is especially important as there are quite some differences in the simulated cloud properties between the models). I assume there are reasons, but why is the vertical resolution different between both models? Throughout the manuscript, the authors point out the importance of entrainment and cloud top cooling for the cloud evolution in both models, however, the vertical resolution is essential in simulating these smaller-scale cloud top processes. Also, the inclusion of hail in RAMS can alter microphysical rates and cloud liquid and ice content, which is not discussed appropriately. Did the authors also perform simulations without including the hail category in RAMS?

4. The authors highlight the importance of Aitken mode particles for cloud sustenance in the title of this work and in the conclusions (line 576), however, in section 3.3.1 it is merely mentioned that Aitken mode particles have a significant impact on cloud droplet mixing ratio for up to 20 cm-3 of accumulation mode particles (RAMS) and 10 (MIMICA), but no statements about cloud sustenance are made. As this is an essential part of the paper, it should be pointed out in the results and be discussed more thoroughly (as mentioned above, also in terms of radiative impact, future implications, seasonal importance etc.).

5. The study has some caveats, which are not addressed at all but would be worth mentioning in a potential discussion section (maybe expand section 5 to a general discussion). Apart from some differing model settings as mentioned in point 3, the CCN are not prognostic, which certainly affects the cloud response to CCN. Similarly, the ice crystal number concentration is set constant, which also has implications for cloud properties in contrast to prognostic INPs such as used for example in Possner et al. (2017), Eirund et al. (2019) and Solomon et al. (2015, 2018). Lastly, secondary ice processes are omitted, however, I would imagine that they could play a role in summertime Arctic clouds as recently shown by Sotiropoulou et al. (2020).

Specific comments

Abstract:

Line 18: I find the expression "large-eddy simulation model" confusing as "simulation" already implies the term model. Maybe consider changing LES model (throughout the manuscript) with simply "LES", "models in LES mode" or similar.

Line 27: Related to my comment 1, it would be good to be more specific here in terms of when and where Aitken mode aerosols matter. You could add something like "for summertime MPCs over pack ice" (implying that this is when Aitken mode aerosols matter the most). If you additionally investigate the radiative response, it would be interesting to mention this here as well.

Introduction:

Line 35: The local lapse rate feedback has also been found to be important for Arctic Amplification, potentially even to be most important (e.g. Stuecker et al., 2018). Please add this here.

Line 40: The Arctic can also be in a persistent cloud-free state, thus consider changing "permanent" with "long-lived".

Line 43: "A layer of liquid is typically present at the top of SMP clouds" Please add a reference.

Line 53: The importance of free tropospheric humidity has also been shown by Solomon et al. (2011,2014) and Loewe et al. (2017).

Line 59: Consider linking these two paragraphs to point out that cloud microphysical properties and thus their radiative effect is for example impacted by aerosols.... (then go into introducing aerosols)

Line 62: There are actually a number of studies that have shown sensitivity of Arctic sea ice clouds to aerosols (e.g., Solomon et al., 2018, Stevens et al., 2018, Eirund et al., 2019).

Line 65: Also here there are more studies (e.g. the studies mentioned above) that have shown a strong impact of CCN changes to the radiative balance at the surface.

Line 88: "indirectly inferred" - this is very vague, what exactly did they show?

Line 89: From this section it is not clear to me what is different south of the ice edge as compared to over pack ice and between the Arctic and the high Arctic. Please be more specific in what previous studies have shown for which conditions and why more research is necessary.

Line 102: This is confusing to me. According to Koehler theory, large particles should always dominate the CCN availability as they activate more easily. Maybe change "even at low total aerosol concentrations" to "low accumulation mode aerosol concentrations"?

Methods:

Line 121: Are both of these models commonly used for simulating Arctic mixed-phase clouds? I have read several studies including MIMICA, but to me the RAMS model is rather uncommon for simulating Arctic clouds, so it would be good to add some references here that have used these models previously for similar studies.

Line 168: Here is one example (in addition to the introduction), where you point out the importance aerosol-cloud interactions for the surface energy budget, which is however never addressed. If you mention it as it is done here, it needs to be analyzed, otherwise please remove.

Line 170: According to the beginning of this section I assume the radiosonde was launched over sea ice, such that the surface conditions in the model are set to sea ice? If this is the case, please explicitly mention.

Line 173: "The cloud base and cloud top were nearly constant during the cloud lifetime (500 and 1000 m, respectively)." This implies to me that the cloud has been observed over a longer time period, however, in Figure 1 it looks like the observations show only one point in time. Please clarify and/or change the layout of Figure 1 (see my comment regarding Figure 1).

Line 196: "The concentrations are chosen to cover typical aerosol size distributions often encountered in the summertime high Arctic (Heintzenberg and Leck, 2012; Leck and Svensson, 2015)." – for both, accumulation and Aitken mode aerosols?

Line 197: Better use "assumed" rather than "considered"?

Line 200: As you performed quite a large set of simulations, if would be helpful for the reader to have an additional table including all simulations and the varying type/number of aerosol and model that could be referred to here. Also simulation AC20_AK20 could be clearly marked as baseline or control simulation.

Line 203: "fluxes were small" - how small (please be specific)? Is a prescribed flux of zero justified?

Same line: Is the choice of 0.844 for surface albedo arbitrary or is there a reference?

Same paragraph: Was there large-scale advection? And how was the roughness length defined? Also I assume the surface condition was set to sea ice? Please add these information to the simulation setup.

Line 210: Based on what conditions was a spin-up of 2 h chosen?

Line 229: Why were these simulations only performed with MIMICA?

Line 232: "relatively ice-free" An ice crystal number concentration of 0 L-1 is completely ice free, not only relatively ice-free. I would remove "relatively".

Results:

Figure 1: As mentioned above, it looks like you are comparing the temporal evolution of the modeled clouds with an observed temporal snapshot (if this is the case). It would be helpful to have more information about the observations (also how are the percentiles derived, are the observations constant in time?) in the text and the Figure caption. Also it would be interesting to know, if the models and the observations cover the same vertical range of the cloud (if these information are available from the observations). You could consider changing the layout of this figure to height on the y-axis and cloud liquid water and ice content over height on the x-axis.

Line 255: It looks to me that RAMS has a higher autoconversion, as also qr is higher (as shown in Figure 2). If this is indeed the case, maybe mention autoconversion as an additional point.

Line 259: Could another reason be the inclusion of hail in RAMS? Does hail maybe increase surface precipitation, which then reduces the overall LWP and IWP in RAMS? Have you tried to switch hail off?

Line 265: Is it possible that the additional rain formation in RAMS stabilizes the cloud and prevents a continuous cloud top rise as seen in MIMICA?

Line 266: Where do these peaks in qi come from? I would expect to see corresponding peaks for example in the radiative cooling rate, which would hint towards enhanced growth by deposition at certain times, but I cannot find any evidence there.

Line 270: Entrainment can also lead to drying (Ackermann et al., 2004) - was there a moist layer present in the observations? Maybe it would be good to show the initial profiles as measured by the radiosonde to see the temperature and specific humidity (or total moisture) vertical distributions? Also is looks like there is enhanced evaporation of cloud droplets just above cloud top (Figure 3a,b), which would hint towards a drier layer overlying the cloud? In this case entrainment would rather dry than moisten the cloud.

Line 278: Is the correlation of condensation with updrafts shown anywhere? Or is this a general statement? In the latter case please add a reference.

Figure 3: Please consider changing the colorscale. I understand that smaller values in MIMICA have to be represented, but for example in Figure 3d or f I cannot see the total magnitude at all.

Line 329: "thins" rather than "shrinks"

Line 334 and Figures 5 and A2: Is this the cooling or the heating rate (such that neg. values indicate a cooling as in Brooks et al. (2017) which is what I assume following your arguments)? Also the differing colorscales are very confusing, especially since the numbers are very small and an additional zero can easily been overseen. Please consider changing the units to K/h or K/d and try to keep the colorscales the same. As the cloud top is essential for the analysis, it might be worth zooming into the cloud top to better resolve the magnitude of the cooling there. You could also add cloud top such as shown in Figure A3 within Figure 5, so the reader can easily identify cooling at the cloud top.

Line 337: In line 253 you say that stronger radiative cooling rates in MIMICA produce a higher LWP, while here you state that the higher qc leads to stronger cooling in MIMICA. Of course it's a feedback where high LWP changes the cloud emissivity, which in turn increases longwave cooling which then

again favors enhanced turbulence and condensation. However, above a certain threshold of approximately 40 g m-2 the sensitivity of longwave cooling to LWP becomes small (e.g. Garret and Zhao, 2006) which is why I assume you point out the difference in cloud top liquid water here. Please be more specific in your line of argumentation of the qc/cloud top cooling feedback and also refer to other studies of Arctic mixed-phase clouds which have investigated this correlation as well.

Line 343: Not only the entrainment of moist air, but also the increase in vertical motions as a result of more turbulence can favor condensation and maintain cloud liquid (Shupe et al., 2008).

Line 347: As mentioned in my point 3, does maybe the different vertical resolution also play a role here as well?

Figure 4: It looks like the high end of the colorscale is never reached, thus consider adjusting it.

Line 380: I see a very strong signal of Aitken mode particles for low number concentrations of accumulation mode particles, which underlines the main message of the manuscript (cloud maintenance for 6 h). Why do you say there no clear trend?

Line 387: Here you say there is a significant impact, while above you write there is no clear trend (see my previous comment). I would agree with this statement and emphasize it more, as it is one of the main messages of the manuscript.

Line 410: why do more Aitken mode particles lead to more turbulence? Because of more latent heat release through condensation? If this is the case, please clearly state it. However, the updrafts for low accumulation mode and different Aitken mode particles look very similar, but this could be due to temporal averaging I assume.

Line 415/Figure A9: I assume Figure A9 shows spatial and temporal statistics? In this case I cannot see a temporal evolution in the updrafts that would determine the temporal evolution of rain. Please make this argument clearer.

Line 418: Previous studies have also shown an impact of CCN on cloud ice and increased LW cooling of CCN-perturbed clouds has been identified as driving force for increased immersion freezing and growth by deposition (Possner et al., 2017, Solomon et al., 2018, Eirund et al., 2019). As the ice crystal number concentrations is fixed, immersion freezing does not play a role here, but I would suspect that LW cooling does also impact growth by deposition and thus qi. This might be worth exploring/mentioning.

Line 426: This is an interesting finding. The effect of CCN changes for different background ice crystal number concentrations/INP concentrations has been studied previously (Stevens et al., 2018, Possner et al., 2017). There, also a smaller CCN impact was found for higher ice crystal number concentrations /INPs, which would agree with your findings and might be worth mentioning.

Supersaturation statistics:

Line 487: Why did you choose a 20 min interval around 6 h of simulation time? From Figure 6 it looks like strongest signals in qc are either in the beginning or towards the end of the simulations (in RAMS); is your choice of time period linked to any criteria?

Line 515: Again, previous studies have also shown an increase in cloud-top radiative cooling in seeded clouds (see my previous comment). Please cite previous studies accordingly.

Qualitative comparison of model results with observational data for the High Arctic:

Line 338/Section 5: This section is very interesting and gives the authors the opportunity to make statements about the relevance of their work in a broader scope. However, this section is relatively short in my opinion and could be expanded. Especially, it would be interesting to know when/where is including Aitken mode particles important and should be considered in modeling studies? Also the authors could expand section 5 to a general discussion of the results and could compare their findings to previous work which investigated the response of Arctic cloud to varying CCN concentrations.

Summary and conclusions:

Line 580: "Aitken mode aerosols have a significant impact on the cloud droplet amount" - All Figures show differences in mixing rations, not number concentrations (hence cloud droplets could also be larger, not necessarily more numerous). Did I miss something or should this be "cloud liquid water"? I agree that the increase in qc most likely results from an increase in Ndrop, but this has not been shown, as far as I can see.

Line 582: Again "higher number of cloud droplets", same comment as above.

References

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Stuecker et al., 2018: Polar amplification dominated by local forcing and feedbacks, Nature Climate Change, 8, 1076-1081, doi: 10.1038/s41558-018-0339-y