

Reply to referee's #1 comments on manuscript:

5 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.

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15 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.

20 We thank the reviewer for his/her careful reading of the manuscript and many constructive comments.

General comments

25 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.

30 Thus, it should also be emphasized more, when and where Aitken mode particles matter – here, 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.

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40 We have now expanded the text on previous studies that have investigated the CCN influence on high Arctic clouds and the seasonal cycle of aerosol particles in this region in Section 1 and 5. We have also contrast areas over the pack ice and open ocean. In this way we pointed out why Aitken mode particles can be particularly important for the summertime low-level SMP clouds over the Arctic pack ice area. Section 5 includes new Subsection: 5.2. *General importance of Aitken mode particles for low-level mixed-phase cloud properties.*

45 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.

50 We thank the reviewer for this important input and have now added a figure that shows the differences in downward LW radiation at the surface between the cases with the same number of accumulation mode particles but with different Aitken mode concentrations (Figure 10). Differences in SW radiation were found to be small and are only discussed in the text. We have also added the surface precipitation figure in the Appendix.

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60 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?

65 Our aim was to use the default setup for both models, as this is what typically would be used for an arbitrary study. For example, modelling studies that use only one model to compare with observations would most likely use the default model setup. The default setup in RAMS is a fixed vertical grid spacing while in MIMICA the default is a variable grid spacing. Nevertheless, we agree with the reviewer that the difference in vertical grid spacing could be an important reason for the simulated differences in cloud properties. Therefore, we ran an additional MIMICA simulation for the baseline case with a fixed vertical grid spacing of 10 m like in RAMS. The test did not show any significant difference in the simulated cloud microphysical properties compared to the MIMICA baseline case with a variable grid spacing. Thus, the difference in resolution at cloud top does not impact our conclusion. The inclusion of hail is also a standard setting in RAMS. We agree that hail generally can change microphysical rates, however, in all RAMS simulations 97.9-99.7% of the ice is present as ice crystals so the riming treatment plays a very small role in these simulations. We have also checked the hail contribution to the total surface precipitation rates and it is 2 order of magnitude less than the contribution from rain. The differences in model setup in terms of vertical resolution and hail are now discussed in the manuscript (Subsect. 2.3).

80 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.).

85 We agree with this point. We have now added a statement about the cloud sustenance in Sect. 3.3.1. In the same section, we have also added a figure that shows the influence of Aitken mode particles on the downward LW radiation at the surface (Fig. 10). The seasonal importance and future implications are discussed in the Sect. 5.2 and 6.

90 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).

100 Please see our reply on point 3 regarding the different model configurations. The number of CCN is actually prognostic in the study as it is calculated from the prescribed aerosol size distributions. However, there is no sink or source of aerosols. We agree with the reviewer that if there would be the sink, then the effect of Aitken mode particles would be most likely even larger since accumulation mode particles are larger in size and thus they would be removed more easily. This is now mentioned in the new version of the manuscript (Subsect. 5.2). We also agree with the reviewer that the prognostic INPs could give a higher sensitivity compared to prescribed ice crystal concentrations. This is now discussed in the Discussion subsection 5.2. The aggregation of ice crystals is actually included in both models. The previous statement that aggregation is omitted in MIMICA was a mistake and has been corrected.

110 Specific comments

Abstract:

115 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.

120 We respectfully disagree with the reviewer on this point and argue that “LES model” can be used. MIMICA and RAMS are models that utilize large-eddy simulation (as a technique). We have thus kept the term “LES model” in the manuscript.

125 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.

130 The first sentence in the abstract was: “The potential importance of Aitken mode particles (diameters ~25–80 nm) for stratiform mixed-phase clouds in the summertime high Arctic has been investigated using two large-eddy simulation models”. We have now added: “...summertime high Arctic (> 80° N) ...” to be more specific what term “high” means.

We have also added that the Aitken mode particles have a significant impact on radiative properties of the cloud (which is shown in the Results section).

135 **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.

140 We agree with the reviewer. The local lapse-rate feedback has been added in parenthesis as one local feedback. The reference is also added.

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

145 Indeed, this was a mistake. “permanent” has been changed with “persistent”.

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

150 References have been added.

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

155 In the previous version of the manuscript, this paragraph was focused on low-level SMP clouds in the high Arctic (> 80° N), which was why we only included references investigating this specific region. However, since the explanation is true in general for SMP clouds in the Arctic, we have modified the paragraph so that it is not only focused on latitudes > 80° and the suggested references have been added. In the paragraph after this one, we focus on the high Arctic region and stick to the corresponding references.

160 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).

This has been changed now.

165 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).

The introduction has now been changed and this sentence has been modified.

170 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.

As we have changed the introduction this sentence is now removed.

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

180 We have changed this. It is now: However, these analyses were not performed for the high Arctic and they did not explicitly investigate the relation between Aitken particles and cloud properties or cloud sustenance. Instead, they focused on the correlation between aerosol particles and cloud droplets.”

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.

185 We have now written about these differences and added corresponding reference.

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"?

190 We agree with the reviewer that the sentence was unclear. We have changed it to: "We initialize the models with a range of aerosol size distributions and explore if Aitken mode particles can help sustain the cloud or if only accumulation mode aerosols control cloud properties (i.e. cloud droplet, rain and ice mixing ratios), even at low accumulation mode concentrations."

195 **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.

200 We have added references for both models. However, even if one of the models had not been specifically designed/used for Arctic mixed-phase clouds we still believe that it would be of interest to show that it generates the same qualitative results as a model that has been used to simulate these clouds.

205 **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.

210 This is a good point. In the new version, we have addressed the importance of Aitken mode particles on radiative fluxes.

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.

215 Yes, the surface conditions were set to sea ice. We have now stated that explicitly, both here and in the *Simulation setup* section. This sentence is now: "During the ice drift, radiosondes were launched **from the ice surface** every 6 h and provided profiles of thermodynamic properties (e.g., pressure, temperature, relative humidity) and wind speeds (cf. Figure 1)."

220 **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).

225 Yes, the cloud was observed over a longer time period as stated in the section 2.2.: "To investigate a case with a quasi-steady-state cloud regime, the simulations are based on a period that was characterized by a persistent, low-level SMP cloud observed from 18 UTC 30 August to 12 UTC 31 August 2008." The observations in Figure 1 (now Figure 2) do not show only one point in time, which was also mentioned in the figure caption: "The retrieved values of LWP and IWP for the **observed** period are shown as 25th, 50th (median) and 75th percentiles." However, to be more clear, we have modified the caption of Fig. 2:" The retrieved values of LWP and IWP for the observed **cloudy period defined in section 2.2 and up to the height of the model domains** are shown as 25th, 50th (median) and 75th percentiles."

230 **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?

235 Yes, for both accumulation and Aitken mode aerosols. The cited studies show the aerosol size distributions in the high Arctic, i.e. include both modes.

Line 197: Better use “assumed” rather than “considered”?

“considered” has been changed with “defined”.

Line 200: As you performed quite a large set of simulations, it 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.

We also thought that a table was a good idea, however, once we made it we did not think it was particularly useful (we have included it as a supplement, just FYI). Therefore, it is not included in the new version of the manuscript.

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

The observed fluxes were usually smaller than 5 W m^{-2} during the ASCOS ice drift period with probability peaks around zero (Tjernström et al., 2012). Previous modelling studies simulating the ASCOS case (e.g., Stevens et al., 2018) have also used zero surface fluxes. We have added this explanation in the new version of the manuscript.

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

We have now added “(cf. Sedlar et al., 2011)” as the reference for the surface albedo value.

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.

There is no large-scale advection and it is now stated in the manuscript. The information about the roughness length and sea-ice surface conditions have also been added.

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

We consider that after 2h a stable cloud is developed. The information has now been added: “The first 2 h are assumed to be a spin-up period and is therefore excluded from the figures and analysis. After the spin-up period, the cloud layer is stable in the baseline simulations”.

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

These simulations have now been done also with RAMS. The results are included in the new version of the manuscript (Subsect. 3.3.1, Figures 11 and 12).

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”.

We agree, “relatively” has been removed.

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.

We have changed the figure caption to: “... The retrieved values of LWP and IWP for the observed **cloudy** period defined in section 2.2 and up to the height of the model domains are shown as 25th, 50th (median) and 75th percentiles...” For more details, please see our reply to the comment *line 173*.

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.

305 The figure shows q_r profiles for the baseline simulation only and indicates slightly more rain in RAMS just below the cloud. The cloud q_r values are quite similar among the baseline cases simulated by the two models. This is now explicitly stated: “Rain mixing ratios are similar for the two models (Fig. 3c and 3d), but RAMS produces slightly more rain below the cloud.”

310 Moreover, looking at the figure A4 where the q_r profiles are presented for all cases we can conclude that there is in general more rain in the cloud in MIMICA than in RAMS. This is especially noticeable when comparing the cases with low accumulation mode particles where RAMS does not produce a stable cloud. MIMICA simulates a thicker cloud in all cases comparing to RAMS, with more turbulence (now added as Figure A6) and more liquid in general, as a consequence of 2-3x higher cooling rates at the top of the cloud (as is stated in the manuscript).

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?

315 The surface precipitation is in general higher in MIMICA than in RAMS, which agrees with higher cloud q_r values produced by MIMICA (the surface precipitation figure is now included in the manuscript, Figure A7). We have also checked the individual contribution from rain, ice and hail to the surface precipitation in RAMS (not shown). The contribution from hail is 2 orders of magnitude smaller than the one from rain and 1 order of magnitude smaller than the one from ice; thus the inclusion of hail in RAMS does not change the overall conclusions. This is now discussed in the manuscript.

320 **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?

325 As we pointed out above, the cases in MIMICA generally have more rain within the cloud and more surface precipitation than the ones in RAMS (Figure A4, as well as added Fig. A7). Based on all analyses we have performed we think that the thicker clouds in MIMICA (with rising cloud tops, stronger turbulence and more liquid in general) are most likely the consequence of the different cloud top cooling rates produced by the two models.

330 **Line 266:** Where do these peaks in q_i 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.

335 The peaks in total ice mixing ratio in MIMICA come from different collection rates of rain by graupel that arise with time (now added as Figure A8). The change in collection rates with time is a consequence of peaks in updrafts and downdrafts (the same applies for all cases, figures are shown for the baseline simulation only (Figure A8)). This has been also added in the new version of the manuscript: “Both models also simulate similar values of total ice mixing ratios, although MIMICA produces a few stronger vertical bands after 6 h of simulation (Fig. 3e and 3f). **This type of pronounced bands is a result of strong collection rates of rain drops by graupel, which appear at different times due to different temporal distributions of updrafts and downdrafts (Figure A8).**”

340 **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 it 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.

345 We agree that the entrainment may also lead to drying depending on the humidity above the cloud. Stratocumulus-topped boundary layers in the high Arctic are usually capped by both temperature and humidity inversions leading to the entrainment of moist air into the boundary layers. This was stated in the introduction in the previous version. However, we agree with the reviewer that we could point out more clearly that this also applies to our simulations. We have included a figure with the radiosonde observations of absolute temperature, potential temperature and specific humidity (Figure 1) where both the temperature and humidity inversions can be observed. The sentence has also been changed to: “The reason is most likely the higher entrainment rates at cloud top in MIMICA (not shown) that bring more water vapor into the cloud from the moist air that is present across the humidity inversion, which caps the cloud-topped boundary layer (Figure 1c).”

355 There is indeed a thin layer of cloud droplet evaporation just above the cloud. However, based on entrainment rates and the humidity profiles (both observed and simulated) we expect positive vapor flux into the cloud layer.

360 **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.

This conclusion was drawn from an analysis of the RAMS results and it is thus not a general statement. We have clarified this by adding “(not shown)”.

365 **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.

This has been changed, we have now added a colorscale for each plot.

370 **Line 329:** “thins” rather than “shrinks”

We have changed “shrinks” with “thins”.

375 **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 be 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.

380 Yes, the figures represent the heating and not the cooling rates. This was a mistake. We thank the reviewer for all suggestions regarding these figures, they have now been changed accordingly (now Fig. 6 and A2).

385 **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.

390 We agree with this point. We do not consider that the difference in the radiative cooling rates between the two models arise from this feedback, it is simply a consequence of the different radiation schemes that the models use. The additional tests we performed and presented in the Appendix support this conclusion.

395 In the new manuscript version, we have explicitly written that the discrepancy in cloud top cooling does not arise from the correlation between LWP and cloud top emissivity and we have cited Garret and Zhao (2006). “For the baseline case, both models simulate a relatively thick cloud (LWP > 40 g m⁻²), which indicates that the differences in the cloud top cooling rates between MIMICA and RAMS do not arise from the difference in simulated LWP (cf., Garrett and Zhao, 2006).” is now added.

400 **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).

405 This sentence has been removed from the new version for other reasons, but we do agree with this point.

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

410 Please see our reply to point 3.

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

415 This has been adjusted.

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?

420 We do think there is an influence of Aitken mode particles in all cases in both models as it was stated in the first
sentence of the paragraph.” Figure 6 shows that adding Aitken mode particles generally increases the amount of
cloud droplet water in both models, i.e. the particles serve as CCN and allow formation of additional cloud
droplets.”. The student t-test confirms it. What we wanted to say is that there is no clear trend in the Aitken mode
425 influence between the RAMS cases with different (and low) accumulation mode concentration, i.e. that is not clear
whether the Aitken particles are more important for the cases with $acc=0\text{ cm}^{-3}$, $acc=3\text{ cm}^{-3}$ or $acc=5\text{ cm}^{-3}$.

We agree with the reviewer that the sentence was confusing. Moreover, we now think the figure shows that the
influence of Aitken mode particles in RAMS generally also becomes less pronounced as the accumulation mode
concentration increases (as it is in MIMICA). This is now changed in the manuscript:” In both models the influence
of smaller particles on cloud droplet mixing ratio thus generally decreases with increasing accumulation mode
430 concentration (Fig. 7). Nevertheless, the differences in cloud droplet mixing ratio are statistically significant for
all pairs of different Aitken mode concentrations in both models, except for the MIMICA pair AC20 AK200 and
AC20 AK20 (according to a student t-test with a 95 % confidence level on the time averages in the cloud layer).
In other words, both models show that Aitken mode particles have a significant impact on the cloud droplet mixing
ratio, at least up to 20 cm^{-3} of accumulation mode particles in RAMS and at least up to 10 cm^{-3} in MIMICA.”.

435 **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.

440 Please see our reply above.

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.

445 In the previous version this statement was written as a possible explanation: ”Despite the higher number of cloud
droplets, an increase in the Aitken mode particle concentration **may lead** to stronger turbulence”. In the new
version, we have added a figure with the cloud-averaged time-mean TKE values (Fig. A6). The figure shows that
there is indeed more turbulence in the cases with increased number of Aitken mode particles.

450 **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.

455 Yes, the statistics were done based on a 20min period for all supersaturated grid boxes in the model domains. We
do realize the time variability in updrafts could not been seen from the figure. We refer now to another figure,
which has been added in the new version (Fig. A8).

460 **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 q_i . This might be worth exploring/mentioning.

465 We agree with the reviewer. We have now explained the relationship between increased LW cooling and the
influence of CCN on ice: “The influence of Aitken mode particles on ice is in general larger in MIMICA than in
RAMS consistent with the stronger cloud top cooling rates simulated by MIMICA that favors the ice formation
through immersion freezing and growth by vapor deposition when the number of CCN increases (Possner et al.
2017; Solomon et al., 2018; Eirund et al., 2019).”.

470 **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.

475 This has been added now: “This result agrees well with previous studies that have investigated the influence of
CCN in mixed-phase clouds with different background INP or ice crystal concentrations (e.g., Possner et al., 2017;
Stevens et al., 2018).”.

Supersaturation statistics:

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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?

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In the RAMS cases with low accumulation mode concentrations ($acc=0,3,5 \text{ cm}^{-3}$), the signals are stronger at the beginning than towards the end due to the cloud dissipation after 6h. However, we consider that 6h, as the middle of the simulation period, can be more representative for the statistics than some earlier times. If we chose a time towards the end of the simulation then there would not be a cloud in the all RAMS cases. The period of 20min is chosen to cover data variability and thus be more representative for the statistics than the output from one time step.

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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.

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We have now cited previous studies.

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

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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.

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We have extended this section to a general discussion and made two subsections, 5.1 *Qualitative comparison of model results with observational data for the High Arctic* and 5.2.

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General importance of Aitken mode particles for low-level mixed-phase cloud properties. In the later one, we have discussed the spatial and seasonal importance of smaller particles.

Summary and conclusions:

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Line 580: "Aitken mode aerosols have a significant impact on the cloud droplet amount" - All Figures show differences in mixing ratios, 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.

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We thank the reviewer for pointing this out. We have now changed it.

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

It is changed.

525

Reply to referee's #2 comments on manuscript:

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Interactive comment on "The importance of Aitken mode aerosol particles for cloud sustenance in the summertime high Arctic: A simulation study supported by observational data" by Ines Bulatovic et al.

Anonymous Referee #2

Received and published: 26 August 2020

535

General comments:

The authors employed two numerical models to perform a series of simulations to study the importance of Aitken mode aerosol particles for cloud sustenance in the summertime high Arctic. The messages in the abstract seem to

540 be clear. After reading through the main text of the paper, I find some interesting results. But I am also confused
by arbitrary model and simulation configurations and overwhelmed by poorly interpreted, disorganized, and
probably unnecessary results. Throughout the manuscript, the authors used a lot of speculations in their reasoning
where solid evidences are expected. The writing also has huge room for improvement. I listed my major and minor
545 concerns as well as suggestions regarding the technical aspects of the manuscript below.

We thank the referee for his/her comments that have helped us improve and clarify the manuscript.

550 Both models utilized in the study have previously been used for simulating Arctic mixed-phase clouds, which is
now explicitly stated in the manuscript (Sect. 2.1). We have on purpose used the default configurations of the two
models, as this is what typically would be used for e.g. a model-observation comparison/evaluation.

We have answered the comments by the reviewer in a point-by-point fashion, revised the manuscript accordingly
and made a thorough effort to provide a clearer and more organized interpretation of the results.

555 One of the co-authors on the study is an English native speaker who has carefully read the manuscript and paid
attention to linguistic mistakes.

Specific comments (major):

560 If I understand correctly, the main idea of this paper is that, for some combinations of model configurations (i.e.,
Aitken mode and accumulation mode aerosol number concentration, aerosol kappa value, and ice number
concentration), the modeled cloud can survive through 12 h, meaning that during this period, the clouds can
maintain sufficient liquid water against processes that depletes liquid water (like subsidence, entrainment of warm
air, losing moisture due to glaciation and precipitation, so on) and generate enough supersaturation to activate the
565 prescribed Aitken mode aerosol. Was the specified aerosol size distribution for each simulation used from the
beginning of the simulation? How important are aerosols during the spin-up? In other words, are the differences
among simulations using the same model and between MIMICA and RAMS due to the activation behavior when
the turbulent motion is very weak? What if all simulations (in each model) begin with a robust cloud and fully-
developed the turbulence, spun-up using same configurations, and then switch to different aerosol number
570 concentrations? I don't think a juicy but non-turbulent cloud is a realistic starting point to test aerosols' impacts
on sustaining clouds or produce results that are relevant to the real world. Please justify this choice.

575 Both models use an initial cloud droplet number concentration of 30 cm^{-3} . This is now clarified in the manuscript
(Sect. 2.3). In other words, both models start with a fully developed cloud, which thereafter is maintained (or not)
based on the prescribed aerosol size distributions and modelled supersaturations. We do believe this type of model
setup is realistic as it roughly represents conditions where a cloud forms close to the marginal sea ice zone and
thereafter is advected in over the sea ice (where aerosol sources are either absent or small).

580 A few other questions related to initialization and spin-up: Is the initial cloud size distribution related to the Aitken
mode and accumulation mode aerosol? For each model, are the liquid water content profiles in all simulations
identical at the beginning? Are all microphysical processes (e.g., all processes related to precipitations) turned on
from the beginning?

585 The profile of cloud water mixing ratio is the same in both models at the beginning of all simulations as it is stated
in the manuscript. All microphysical processes are active at the beginning of the simulations, which is now also
stated (please see Sect. 2.3).

590 It is worthwhile to be more specific about the activation of aerosol particles in MIMICA and RAMS as configured
for this study. It seems that the activation scheme in MIMICA is identical to the one used to generate Fig. A10.
What about in RAMS? Are the ss in Fig. 10 and Fig. 11 same as those used in the activation scheme in the models?

595 Figure A10 (now Fig. A12) shows theoretical critical supersaturation values as a function of particle dry diameter
for a range of kappa values, where kappa is a measure of the hygroscopicity of a multi-component or single-
component aerosol particle (Petters and Kreidenweis, 2007). The activation schemes in both models use this theory
for cloud condensation nuclei activation as stated in the Sect. 2.1. The reason why look-up tables are used in
RAMS is only due to computational efficiency.

The supersaturation (ss) statistics shown in Fig. 10 and Fig. 11 (now Fig. 14 and Fig. 15) are directly derived from
the model output, i.e. the ss values shown are indeed used to activate aerosols in the two models. The critical dry

600 diameters shown in the figures are calculated for the ss values (i.e. for 75th and 99th ss percentiles) obtained from the models, based on the relationship presented in figure A12 and explained in detail in Petters and Kreidenweis (2007). We have not made any changes in the manuscript as all this information was already available.

605 It seems that the authors tried to use an observed sounding to set up the baseline simulations, and then perform sensitivity tests on top of that. However, the authors did not provide enough details (for example, dedicated figures) for readers to understand the case. Please consider showing some details. I found the ASCOS sounding available from <https://bolin.su.se/data/ascos-radiosoundings>. Did the authors use the original 0535 UTC 31 August 2008 sounding from this archive? Or an idealized version of it? The authors used the observed CCN to justify the use of AC20_AK20 as the baseline simulation. However, the cloud layer in the aforementioned sounding seems to be decoupled from the surface. If this is the case, does it still make sense to use surface measurement to determine the base case? The authors mentioned a few times that Arctic stratocumulus may entrain moist air from above the cloud top, but provided no evidence. (Whether other studies showed entrainment of moisture from above the cloud could happen is irrelevant to this study.) Initial sounding (together with profiles from the middle of the simulations) can be used to show whether there is moist layer above the cloud top.

615 We agree with the reviewer that it is useful to show the radiosonde observations and have therefore added the new Figure 1. We have indeed used the original soundings from 0535 UTC 31 August 2008, as is stated in the manuscript. In Figure 1, we also display the simulated profiles after 6h of simulation. Showing the initial profiles does not make sense as these are the same as the radiosonde observations.

620 There were no other CCN measurements available from the ASCOS campaign apart from those obtained from a CCN counter that was situated on *Oden*. However, we do agree with the reviewer that the CCN concentrations could be different within (or just below) the cloud due to the decoupled boundary layer. The sentence: “Since the surface boundary layer typically was decoupled from the turbulent layer associated with a cloud (Tjernström et al., 2012), it is however not certain if the CCN concentrations measured at the ship were representative for the cloud layer (cf. also observed vertical profiles of particle concentrations in Igel et al., 2017, Figure 1).” has been added in Section 2.2.

625 Initial profiles of absolute temperature, potential temperature and specific humidity (Figure 1) show that the simulated case was characterized by both a temperature and humidity inversion.

630 Choice of model configurations, shared simulation setup, and experiment design are perplexing and arbitrary. Why were the vertical resolutions different, especially as the authors suspected that the different vertical resolutions may be the source of some discrepancies between the simulation results from the two models (e.g., L254). Why were the microphysics in RAMS so much more complicated (even with hail turned on) than in MIMICA? Why was aggregation turned off due to low ice number concentration, but why only turned off for MIMICA? Why was MIMICA used for ice number concentration sensitivity test while certain ice-related budget terms are not available from it (L289)?

635 As we briefly mentioned above, we wanted to use the models in their standard configurations and examine the similarities/differences in the simulated results. The default setup in RAMS is a fixed vertical grid spacing while in MIMICA the default grid spacing is variable. We agree that the difference in grid spacing could be a source of discrepancies and have therefore run an additional MIMICA baseline simulation with a fixed vertical grid spacing of 10 m as in RAMS. The test did not show any significant differences in the simulated cloud microphysical properties compared to the MIMICA baseline case with a variable grid spacing. This is now explained in the new version of the manuscript (Sect. 2.3).

640 It is not completely clear to us why the reviewer considers that the microphysics in RAMS is much more complicated compared to MIMICA. We agree that the inclusion of hail could change the microphysical rates for e.g. a deep convective cloud, but for an Arctic stratocumulus cloud the hail production is in general very small. Indeed, in all RAMS simulations 97.9-99.7% of the ice is present as ice crystals so the riming treatment plays a minor role in the simulations. We have also checked the contribution of hail to the surface precipitation. It is 2 orders of magnitude smaller than the contribution from rain. This information is now added in the manuscript (please see Sect. 2.3). The aggregation process is actually switched on in both models, the statement that it was turned off in MIMICA was a mistake and has been corrected.

645 We agree with the reviewer that the ice tests could have been performed with both models and have now made the corresponding simulations. We present results from both MIMICA and RAMS in the new version of the manuscript and we find that the results are consistent.

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Much of the results regarding rain and ice are only superficially described and discussed, with no obvious connection to the main goal of the paper, and sometimes contain errors. A few examples are provided here.

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L277: “The pockets of condensation and evaporation present in the main cloud layer are well-correlated with updrafts and downdrafts and they tend to cancel each other in the mean. This is why the average condensation rate in the main cloud is of the same order of magnitude as the one in the sub-cloud layer.” Totally lost.

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We have clarified the sentence. RAMS produces a sub-cloud condensation layer (as Fig. 4b shows) and the sentence explains why this layer is present and so pronounced. The relatively high mean value is a result of infrequent but strong condensation due to sub-cloud convection simulated in RAMS.

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L415: “The presence of both positive and negative differences with time is a result of differences in cloud dynamics with different distributions of updrafts and downdrafts with time that govern the rain production in the cloud (cf. Fig. A9).” But there is nothing about “distributions with time” in Fig. A9.

680

We agree with the reviewer that the Fig. A9 (Now Fig. A11) was not a good choice to refer to. We now instead refer to Fig. A8, which shows that there are different distributions of updrafts and downdrafts with time (Fig. A8b,c). The figure also shows that the temporal evolution of updrafts and downdrafts is well correlated with the evolution of the collection rate of rain drops by graupel (Fig. A8a) and therefore also with the rain budget. Results are only presented for the baseline simulation, but the results are qualitatively the same for all simulations.

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L421: “change in the Aitken mode particle number concentration results in that maximum updrafts are reached at somewhat different times”, what is the significance of this?

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The sentence has been rephrased in the new version of the manuscript.

L422: “Differences are in general greater in MIMICA than in RAMS since there is a slightly more total ice in MIMICA”. Does “since” mean “because” here? Does greater total ice water content (or path?) have to lead to greater differences? Other than the “entrainment of moisture aloft”, here are a few additional examples of unacceptable speculation.

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We have now changed the reasoning for the observed difference in the Aitken mode influence on total ice mixing ratio between the two models. Previous studies have shown that a pronounced CCN influence on ice in CCN-perturbed clouds can be related to stronger LW cloud top cooling (e.g., Possner et al., 2017, Solomon et al., 2018, Eirund et al., 2019), which is present in MIMICA and not in RAMS. Please see the Subsection 3.3.1 for more details.

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L288-289, “Examining the total ice deposition/sublimation rates would most likely lead to similar rates between the two models”. (BTW, “The ice crystal deposition and sublimation rates are higher in RAMS than in MIMICA since the two models partition the total ice deposition differently among ice hydrometeor categories” is also just speculation, isn’t it?)

705

The sentence in the brackets is not a speculation. We have indeed analysed the (available) deposition rates for all ice species in both models and concluded that they differ between the same ice hydrometeor categories in the two models. The sentence has been reformulated to:” The ice crystal deposition and sublimation rates are higher in RAMS than in MIMICA since the two models partition the total ice deposition differently among ice hydrometeor categories (not shown).”

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“Examining the total ice deposition/sublimation rates would most likely lead to similar rates between the two models” is on the other hand something that we cannot be sure about and we therefore removed this sentence from the new version of the manuscript.

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L411-412: “an increase in the Aitken mode particle concentration may lead to stronger turbulence and more cloud liquid water production”. Turbulence intensity and cloud liquid water budget can be diagnosed. It does not make sense to speculate (“may lead to”).

This is a good suggestion and we have now included a figure of the time-mean, resolved TKE within the cloud layer (Fig. A6). In both models, the TKE is higher in cases with a higher number of Aitken mode particles (comparing the pairs of simulations with the same number of accumulation mode particles). The relation between

720 the cloud-averaged TKE and cloud water mixing ratio is obtained by comparing Fig. 5 and Fig. A6. The text has been changed accordingly.

Minor comments:

725 Contents in Sections 2.1 and 2.3 are not clearly separately. For example, why are number of model grid points and resolution introduced in Section 2.1 but domain size in Section 2.3?

We agree that it is more suitable to provide this information in the *Simulation setup* section and have moved it there. Otherwise, we believe that the contents of Sections 2.1 and 2.3 are clearly separated.

730 Please clarify a few model or simulation configurations issues: Is it correct that the time step is 2 s for MIMICA? What about RAMS? The terminal speed is only introduced for MIMICA, what about RAMS? What are the references for the terminal speed formulas used? Is the 0.2 L-1 ice number concentration based on ASCOS observations? Need reference. Why is divergence set to 1.5E-6 s-1? Is the same value used for all model layers?

735 It is correct that the time step in MIMICA is ~2s as stated in the manuscript. In RAMS, the time step is 1.5s and this is now explained (Sect. 2.1).

Information about the terminal fall speed in RAMS as well as the references for both models have also been added (Sect. 2.1).

740 We have added the Stevens et al. (2018) study as a reference for the ice crystal number concentration (Sect. 2.3).

745 In both models, the large-scale divergence is constant in the whole model domain (at all model levels) and the value is chosen to produce a stable cloud layer (cf. Stevens et al., 2018). The explanation together with the reference has now been added in the text (Sect. 2.3).

L130: Is the wet deposition calculated for any tracers in either model?

No, this has not been included as stated in the Section 2.3 (i.e. the aerosol size distributions are prescribed).

750 **L346: Since they are simplified LW cooling calculation that only depends on LWC, why not using same formula for both MIMICA and RAMS?**

755 As mentioned before, we wanted to use models with their standard configurations. Using two different models allows us to evaluate if the results are dependent on the details of a specific model or if we can draw more general conclusions. The additional tests that we have done with e.g. the simplified LW cooling calculations (in MIMICA and RAMS) or a fixed vertical grid spacing (in MIMICA) are meant to indicate which differences in the model configurations lead to the discrepancy in the simulated results. We think this information could be useful for the future modelling studies.

760 **L349: “Stable cloud base”, do you mean “steady”?**

This sentence has now been removed from other reasons.

765 **L383: “Fig. 5”, should be Fig. 6?**

Yes, this was a mistake in typing. However, this paragraph has been modified in the new version of the manuscript and the sentence has been removed.

770 **Examples of poor writing:**

L265: “total ice”, do you mean “total ice mixing ratio”? There are a few “total ice” throughout the text.

775 We agree that “total ice mixing ratio” is more precise than “total ice” and have changed it in that place. However, we did not find any other example of incorrect use of the “total ice” term throughout the text. For instance, in the sentence: “The ice crystal deposition and sublimation rates are higher in RAMS than in MIMICA since the two models partition **the total ice deposition** differently among ice hydrometeor categories (not shown).” or in the sentence: “Examining **the total ice deposition/sublimation rates** would most likely lead to similar rates between

780 the two models, but these rates are not available in MIMICA”, the term “total ice” is used as a possessive adjective and not as a noun. The models partition the (total) ice differently and have different (total) ice deposition/sublimation rates. The simulated (total) ice is then expressed with a mixing ratio variable.

L377: “available water vapor” what is this?

785 We have modified the sentence to: ”A higher number of cloud droplets decreases the maximum supersaturation and the amount of water vapor in the cloud available for activation of smaller particles.”

Suggestions on technical issues:

790 Please consider marking cloud top and base whenever relevant.

We have added the cloud top and cloud base heights in all figures where it could be done.

795 Colors are saturated in some figures, e.g., Fig. 3f. Please adjust.

We have adjusted it.

L292: “from the different model descriptions”, should be “configurations”?

800 “model descriptions” has been changed with “model configurations”.

805 **List of all relevant changes:**

810 We have performed all additional simulations that the referees asked for, e.g. the RAMS simulations with different background ice crystal concentrations as well as the additional MIMICA simulation with constant vertical grid spacing, which served to test the sensitivity of the simulated results on different vertical resolutions present in the model default configurations.

815 We have done additional analyses to answer some of the referees’ comments, e.g. analysed the contribution of hail to the surface precipitation in the RAMS simulations.

820 We have added new figures suggested by referees, i.e. the figures of time-mean cloud-averaged turbulent kinetic energy, time-mean surface precipitation, temporal evolution of updrafts and downdrafts as well as the figure that shows the influence of the Aitken mode particles on longwave radiative fluxes at the surface. We have also discussed all of this in the new version of the manuscript.

All the figures for which the referees had improving suggestions have been changed accordingly.

825 The text in the new version of the manuscript has been modified according to the referees’ comments to improve the explanations.

The additional references suggested by the referees are also included in the revised version.

For details, please see our replies to referee’s comments.

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The importance of Aitken mode aerosol particles for cloud sustenance in the summertime high Arctic: A simulation study supported by observational data

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Abstract. The potential importance of Aitken mode particles (diameters ~25–80 nm) for ~~stratiform~~ stratiform mixed-phase clouds in the summertime high Arctic ($> 80^\circ \text{N}$) has been investigated using two large-eddy simulation models. We find that in both models Aitken mode particles significantly affect the simulated ~~cloud~~ microphysical and radiative ~~cloud~~ properties of the cloud and can help sustain the cloud when accumulation mode concentrations are low ($< 10\text{--}20 \text{ cm}^{-3}$), even when the particles have low hygroscopicity (hygroscopicity parameter $\kappa=0.1$). However, the influence of the Aitken mode decreases if the overall liquid water content of the cloud is low, either due to a higher ice fraction or due to low radiative cooling rates. ~~An~~ An analysis of the simulated supersaturation (ss) statistics shows that the ss frequently reaches 0.5% and sometimes even exceeds 1%, which confirms that Aitken mode particles can be activated. The modelling results are in qualitative agreement with observations of the Hoppel minimum obtained from four different expeditions in the high Arctic. Our findings highlight the importance of better understanding Aitken mode particle formation, chemical properties and emissions, in particular in clean environments such as the high Arctic.

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1 Introduction

The Arctic region is experiencing a rapid increase in surface temperature that is substantially larger than the global average increase (Holland and Bitz, 2003; Hartmann et al., 2013). The enhanced Arctic warming, known as the Arctic amplification, is a result of ~~remote forcings (which modify heat and moisture transport from lower latitudes) as well as~~ local drivers and feedbacks (e.g., local aerosol sources, cloud and ice-albedo feedbacks, ~~lapse-rate feedback~~; Serreze and Barry, 2011; [Stuecker et al., 2018](#); [Stjern et al., 2019](#)) ~~as well as remote forcings (which modify heat and moisture transport from lower latitudes)~~. Changes in the dynamical and microphysical properties of clouds are central to local feedbacks (Curry et al., 1996; Garrett et al., 2009; Kay et al., 2011) due to the strong impact of the clouds on the surface energy budget (Curry and Ebert, 1992; Shupe and Intrieri, 2004) and subsequent sea-ice growth (Kay and Gettelman, 2009; Kay et al., 2011; Tjernström et al., 2015).

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~~In the high Arctic (north of 80°N),~~ Low-level, stratiform mixed-phase (SMP) clouds ~~are persistent/permanent and frequent/b ubiquitous in the Arctic~~ (Shupe et al., 2006; Shupe et al., 2013). Despite the presence of liquid and ice in the same volume and a continuous sink of the liquid phase through ice growth and precipitation, these clouds may persist for several days (Shupe et al., 2006). A layer of liquid is typically present at the top of SMP clouds (e.g., [Shupe et al., 2006](#); [Morrison et al., 2012](#)). Maintenance of this layer is critical for sustaining longwave emission and ensuing cooling at the cloud top (e.g., Persson et al., 2017; Dimitrellos et al., 2020), which enhances a buoyancy-driven turbulent mixing in a layer within and below ~~associated with~~ the cloud (e.g., Tjernström et al., 2005). The turbulence further influences cloud liquid growth. Strong overturning means strong updrafts that allow efficient condensation of vapor onto cloud droplets. It also leads to higher entrainment rates at the cloud top (e.g., Tjernström, 2007). A peculiar feature of the high-Arctic region is that the specific humidity frequently increases over the inversion layer that caps the low-level SMP clouds (Sedlar et al., 2012; Shupe et al., 2013). Entrainment may thus bring more vapor into the cloud and moisten the boundary layer ([Solomon et al., 2011](#); Tjernström et al., 2012; [Solomon et al., 2014](#); [Loewe et al., 2017](#)~~hupe et al., 2013~~). This and other conditions specific for boundary layers in the high-Arctic allow liquid water to persist, and thereby also prevent quick cloud glaciation, despite an opposing effect of ice growth within the cloud (e.g., Morrison et al., 2012).

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895 In the high Arctic (north of 80° N), in contrast to low-level clouds at lower latitudes, SMP clouds in the high Arctic have a net warming positive effect on the surface energy budget during most of the year. Due to a low amount of solar radiation in this region, the warming induced by cloud longwave emission towards the surface is generally larger than the cooling effect due to reflection of solar radiation. However, during the peak-melt season at the end of the summer, high Arctic SMP clouds can have a net cooling effect on the surface energy budget and thereby influence the surface temperature and timing of the autumn freeze-up (e.g., Intrieri et al., 2002; Shupe and Intrieri, 2004; Tjernström et al., 2014). Aerosol particles influence the radiative effect of clouds as they act as cloud condensation nuclei (CCN) and ice nucleating particles (INP) and affect the microphysical and optical properties of clouds (referred to as aerosol indirect effects). Due to the generally pristine conditions in the high Arctic (e.g., Bigg and Leck, 2001), the clouds in this region can be particularly sensitive to the aerosol perturbations.

900 ~~Aerosol particles can act as cloud condensation nuclei (CCN) and ice nucleating particles (INP), which affect the microphysical and optical properties of clouds (referred to as aerosol indirect effects). Previous modelling studies of high Arctic clouds (e.g., Birch et al., 2012; Loewe et al., 2017; Stevens et al., 2018) have indeed shown that the cloud liquid water content and cloud radiative properties are highly sensitive to the concentration of CCN at low CCN (referred to as the tenuous cloud regime). Clouds in the Arctic, especially those over the pack ice in the summertime, appear to be particularly sensitive to perturbations. Therefore, there is in the aerosol population due to the generally pristine conditions (Bigg and Leck, 2001). Droplet number concentrations are often low (Leck and Svensson, 2015), and thus even small changes in the availability of CCN may have a critical impact on the radiative fluxes and surface energy balance (Garrett et al., 2002; Lubin and Vogelmann, 2006; Mauritsen et al., 2011). Therefore, there is a need to better understand sources and sinks of high Arctic aerosols, the seasonal variability as well as their chemical composition, physical characteristics and potential effects on cloud formation. However, due to the harsh conditions, measurements are sparse and generally limited to summertime. Overall, the annual cycle of aerosol particle concentrations in the whole Arctic is characterized by transport of anthropogenic emissions from lower latitudes during the winter season with a peak in April (known as the Arctic haze), relatively pristine conditions during the summertime, and a minimum in the fall (Heintzenberg and Leck, 1994; Tunved et al. 2013; Freud et al., 2017). When long-range transport of aerosols over the pack ice is small, as in summer, the surface number concentrations of accumulation mode particles (sizes typically 80–500 nm; Covert et al., 1996) in the high Arctic are generally below 100 cm⁻³ and occasionally below 1 cm⁻³ (Bigg et al., 1996; Mauritsen et al., 2011; Heintzenberg et al., 2015; Leck and Svensson, 2015). During this time of the year, marine biological activity can provide a source of small, airborne particles, adding to the mass and number of Aitken mode particles (sizes typically 25–80 nm; Covert et al., 1996) (Leck and Bigg, 2005a; Heintzenberg and Leck, 2012; Karl et al., 2013; Heintzenberg et al., 2015).~~

925 ~~the surface number concentrations of accumulation mode particles (sizes typically 80–500 nm; Covert et al., 1996) are generally below 100 cm⁻³ and occasionally below 1 cm⁻³ (Bigg et al., 1996; Mauritsen et al., 2011; Heintzenberg et al., 2015; Leck and Svensson, 2015). When long-range transport of aerosols over the pack ice is small, as in summer, the surface number concentrations of accumulation mode particles (sizes typically 80–500 nm; Covert et al., 1996) are generally below 100 cm⁻³ and occasionally below 1 cm⁻³ (Bigg et al., 1996; Mauritsen et al., 2011; Heintzenberg et al., 2015; Leck and Svensson, 2015). During this time of the year, marine biological activity could provide a source of small, airborne particles, adding to the mass and number of the Aitken mode particles (sizes typically 25–80 nm; Covert et al., 1996) (Leck and Bigg, 2005a; Heintzenberg and Leck, 2012; Karl et al., 2013; Heintzenberg et al., 2015).~~

935 ~~The ability of an aerosol particle to act as a CCN depends upon multiple factors, such as its size and chemical composition (Köhler, 1936), surface tension (e.g., Ovadnevaite et al., 2017; Lowe et al., 2019) and the ambient relative humidity (e.g., Rastak et al., 2017). A larger maximum supersaturation within an air parcel allows smaller and less hygroscopic particles potentially to act as CCN (Köhler, 1936; Petters and Kreidenweis, 2007). On the other hand, the maximum supersaturation is also dependent on the relative abundance of particles, in particular the number of water soluble accumulation or coarse mode particles as they easily act as CCN and subsequently take up water when they grow (e.g., Ghan et al., 1997). Therefore, typical CCN sizes differ among the environments with different aerosol size distributions, compositions and supersaturation values (Seinfeld and Pandis, 2006). In general, water soluble particles within the accumulation mode constitute the largest source of atmospheric CCN (Seinfeld and Pandis, 2006). However, in the summertime Arctic, a relatively low condensation sink of water vapor due to the low number of accumulation mode particles may lead to relatively large maximum supersaturations that could allow Aitken mode particles (that are typically more abundant) to act as CCN. The previous studies that have analyzed the effect of CCN in the tenuous cloud regime have not distinguished between the aerosol particles of different sizes and properties that can have different impacts on clouds. Recent observations for the summertime Arctic region south of the ice edge, which focus or include summertime season, have indeed suggested that particles with diameters below 50 nm can be CCN-active (Willis et al., 2016; Kecorius et al., 2019; Koike et al., 2019). However, these analyses studies were not performed for focused on the high Arctic and they did not explicitly investigate the relation between Aitken particles and cloud properties or cloud sustenance.~~

955 ~~Instead, they focused on the correlation between aerosol particles and cloud droplets. Furthermore, and they only indirectly inferred the potential importance of Aitken mode particles as CCN. The environment situation over the pack ice is unique with fewer aerosol sources and different surface conditions compared to the open ocean (e.g., Leck and Svensson, 2015). This could lead to an even stronger influence of Aitken mode particles than south of the ice edge. and the knowledge that we have from other regions south of the ice edge may not be directly applicable.~~ Model simulations by Christiansen et al. (2020) have indicated that Aitken mode particles can influence high Arctic cloud properties, but ~~their these~~ simulations only considered extreme conditions with no accumulation mode aerosols present in the atmosphere.

960 Summarizing, we know that high Arctic summertime SMP clouds over the pack ice are governed by a complex interplay between dynamics, cloud microphysics and aerosols and that they strongly influence climate, however, there are still many uncertainties regarding these clouds. One knowledge gap is if and under which conditions Aitken mode particles become CCN-active in this environment and how these particles then may affect the microphysical properties of the clouds. In this study, we therefore employ two different large-eddy simulation (LES) models to simulate a relatively long-lived summertime cloud observed in the high Arctic during the ASCOS campaign (Tjernström et al., 2014). During the campaign, measurements often showed low concentrations of accumulation mode particles while the concentration of Aitken mode particles was relatively high (Leck and Svensson, 2015). We initialize the models with a range of aerosol size distributions and explore if Aitken mode particles can help sustain the cloud, or if only accumulation mode aerosols ~~dominate the control of~~ cloud properties (i.e. cloud droplet, rain and ice mixing ratios), even at low accumulation mode at low total aerosol concentrations. We also analyze the maximum supersaturations simulated by the two models and calculate the corresponding threshold diameters of aerosol activation. The engagement of two different models allows us to evaluate if the results are dependent on the details of a specific model or if we can draw more general conclusions. Finally, we introduce the statistics of the aerosol size distributions (Heintzenberg and Leck, 2012) observed during the summers of four different high Arctic campaigns that took place in 1991, 1996, 2001 and 2008 year (Leck et al., 1996; Leck et al., 2001; Leck et al., 2004; Tjernström et al., 2014) and compare them with the simulated results. The general conclusions are provided at the end of the study.

980 2 Method

985 2.1 Models

990 The simulations were performed using two models. MIMICA (the MISU MIT Cloud-Aerosol Model) is an LES model that has been successfully used for simulating Arctic mixed-phase clouds (e.g., Ovchinnikov et al., 2014; Savre et al., 2015; Igel et al., 2017; Stevens et al., 2018, Christiansen et al., 2020). ~~The model and~~ solves the equations for a non-hydrostatic, anelastic atmospheric system; a full description of the model can be found in (Savre et al. (2014)). ~~A~~ The model uses a two-moment bulk microphysics scheme (Seifert and Beheng, 2001) is used to calculate the prognostic variables (i.e. mass mixing ratio and number concentration) of five different hydrometeor types considered, namely cloud droplets, raindrops, cloud ice, graupel and snow. All hydrometeor categories have mass distributions in the form of regular gamma functions. Autoconversion and self-collection of liquid particles are also calculated as described in Seifert and Beheng (2001). A pseudo-analytic method is used to model the supersaturation, with the integration of condensation/evaporation at the model time step of ~2 s (Morrison and Grabowski, 2008). The terminal fall speed of the hydrometeors is calculated using a simple power law of the diameter of the particle, which determines the wet deposition (Pruppacher and Klett, 1997). ~~To~~ represent an aerosol population of different particle sizes and chemical compositions, MIMICA includes a two-moment aerosol module (Ekman et al., 2006). All aerosol modes are described with lognormal distributions. In the model, aerosols can act as cloud condensation nuclei following kappa-Köhler theory (Petters and Kreidenweis, 2007), but not as ice nuclei. The number concentration of ice crystals is prescribed ~~to~~ 0.2 L^{-1} and ~~it is~~ kept quasi-constant during the simulations (Ovchinnikov et al., 2011, 2014). This parameterization mimics immersion freezing, i.e. ice can only form if there is supercooled water present. Aggregation of ice crystals is permitted in the model. ~~Secondary ice production and aggregation of ice crystals are omitted in MIMICA.~~ The radiative transfer is calculated following a four-stream radiative transfer solver (Fu and Liou, 1993), which includes 6 bands for shortwave and 12 bands for longwave radiation. ~~The model domain is three dimensional and it is defined by $96 \times 96 \times 128$ grid points. In the horizontal direction, there is a fixed grid distance between the grids of 62.5 m. In the vertical direction, the grid is variable with the highest resolution (7.5 m) at the surface and in the cloud layer.~~

The second model used is the Regional Atmospheric Modeling System (RAMS; Cotton et al., 2003), which has also been successfully used in studies of Arctic mixed-phase clouds (e.g., Avramov and Harrington, 2010; Ovchinnikov et al., 2014). RAMS is a flexible model that is most commonly used for cloud-resolving and large eddy simulations. It uses a two-moment bin-emulating bulk microphysics scheme to predict the mass and number mixing ratios of liquid and ice hydrometeor species (Meyers et al., 1997; Saleeby and van den Heever, 2013). In this study, six species are used, namely, cloud droplets, raindrops, cloud ice, snow, graupel and hail. RAMS typically uses two cloud ice species, but only one was used in this study ~~with a prescribed concentration of 0.2 L^{-1}~~ as was done in MIMICA. Collision-coalescence of liquid drops is done through the use of lookup tables that are generated by solving the stochastic collection equation (Feingold et al., 1997). Condensation depends explicitly on the hydrometeor properties and allows for supersaturation at the end of the time step of 1.5 s (Walko et al., 2000). The terminal fall speed of the hydrometeors is calculated based on piecewise power laws (Mitchell, 1996). RAMS also includes a user-defined number of lognormal aerosol distributions. Aerosol particles act as CCN and are activated using additional look-up tables generated from an offline parcel model based on kappa-Köhler theory (Saleeby and van den Heever, 2013). A Unlike MIMICA, aggregation of ice crystals is permitted. Radiative transfer is calculated following Harrington (1997). ~~The model domain uses the same horizontal specification as MIMICA, but vertical grid spacing is kept constant at 10 m.~~

2.2 Overview of the simulated case: The ASCOS campaign

The Arctic Summer Cloud Ocean Study (ASCOS) campaign took place in the summer of 2008 onboard the Swedish icebreaker *Oden*, and including a three-week ice drift with enhanced meteorology measurements when *Oden* was anchored to a large ice floe slightly north of 87° N . A full description of the expedition can be found in Tjernström et al. (2014). This campaign has so far been one of the most extensive studies in the central Arctic focusing on the atmosphere, clouds and aerosol properties and their linkages to the microbiological life in the upper ocean. To investigate a case with a quasi-steady-state cloud regime, the simulations are based on a period that was characterized by a persistent, low-level SMP cloud observed from 18 UTC 30 August to 12 UTC 31 August 2008. The period represents one of the last days of the ice drift episode, which took place from 12 August to 2 September 2008. During this period, the number concentration of accumulation mode particles was relatively low (Leck and Svensson, 2015). Therefore, a change in the aerosol population could be particularly important for inducing cloud perturbations that may affect the surface energy budget. During the ice drift, radiosondes were launched from the ice surface every 6 h and provided profiles of thermodynamic properties (e.g., pressure, temperature, relative humidity) and wind speeds (cf. Figure 1). The one from ~~approximately 05:356~~ UTC 31 August 2008 was representative of the conditions observed during the ~~whole~~ stratocumulus period and used to initialize the simulations. Cloud properties and thermodynamic characteristics of the atmosphere were monitored with surface-based remote sensing instruments (Shupe et al., 2013). The cloud base and cloud top were nearly constant during the cloud lifetime (500 and 1000 m, respectively). Retrievals of liquid water path (LWP) were made from the 23 and 30 GHz microwave radiometer measurements (Sedlar and Shupe, 2014). The combination of different sensors was used to estimate ice water path (IWP; Shupe et al., 2008). ~~Retrievals of liquid water path (LWP) were made from the 23 and 30 GHz microwave radiometer measurements (Sedlar and Shupe, 2014).~~ The observed LWP uncertainty was around $\sim 25 \text{ g m}^{-2}$ while the uncertainty in the ~~ice water path (IWP)~~ was about a factor of two (Shupe et al., 2008; Birch et al., 2012). A CCN counter that was situated on *Oden*, at 25 m above the sea surface, measured a mean CCN concentration of about 25 cm^{-3} at a supersaturation of 0.2 % during the period of the ice drift (Martin et al., 2011; Leck and Svensson, 2015). The ship and the inlets were facing the wind so that local pollution from the ship was avoided. Furthermore ~~Additionally~~, a pollution controller was used to prevent direct contamination from the ship and the main pumps were turned off whenever the conditions for a clean environment were not completely satisfied (details on the pollution control system can be found in Leck et al., 2001 and in Tjernström et al., 2014). Since the surface boundary layer typically was decoupled from the turbulent layer associated with a cloud (Tjernström et al., 2012), it is however not certain if the CCN concentrations measured at the ship were representative for the cloud layer (cf. also observed vertical profiles of particle concentrations in Igel et al., 2017, Figure 1).

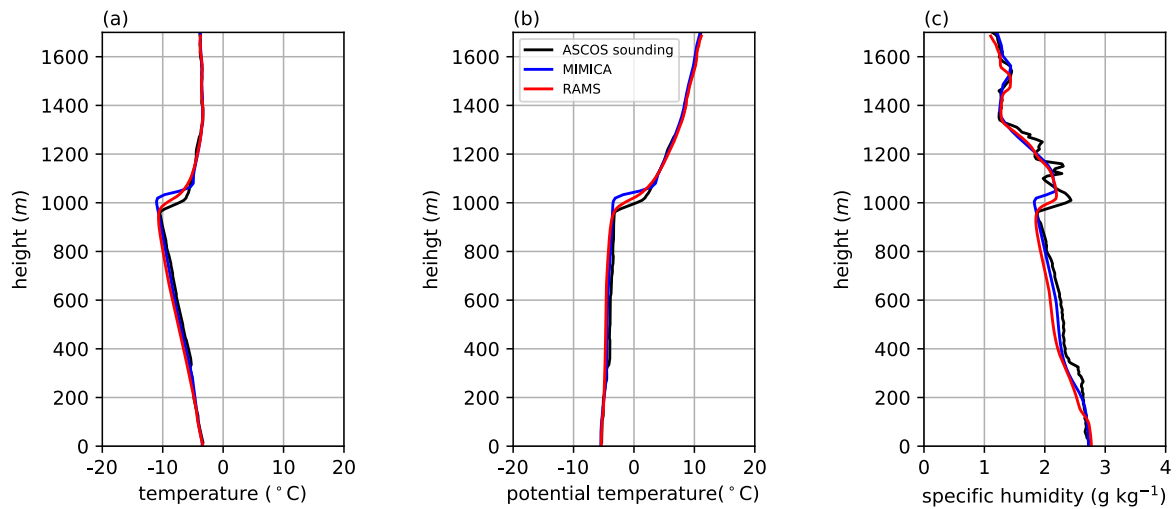


Figure 1: Radiosonde observations from 05:35 UTC 31 August 2008 of (a) absolute temperature (b) potential temperature and (c) specific humidity. The corresponding simulated profiles from MIMICA and RAMS are presented for the middle of the simulation period (6h of simulation time).

2.3 Simulation setup

We performed simulations with different prescribed aerosol size distributions to investigate the influence of Aitken mode particles on cloud properties, and the how this influence dependences of this influence on the background concentration of accumulation mode aerosols. ~~we have performed simulations with different prescribed aerosol size distributions~~ For both models. This means that there is no sink or source of aerosols within the model domain during the ~~e whole~~ simulations for both models. We conduct two sets of simulations with two different background concentrations of the Aitken mode particles (20 and 200 cm^{-3}). Each set contains five cases with different levels of accumulation mode particles (0, 3, 5, 10 and 20 cm^{-3}), i.e. 10 simulations in total. All particle concentrations are assumed to be constant with height. The simulations are named by a combination of two numbers, where the first number refers to the accumulation mode and the second to the Aitken mode number concentration (e.g., the case with 0 cm^{-3} of accumulation and 20 cm^{-3} of Aitken mode particles is referred to as the AC0_AK20 simulation). The concentrations are chosen to cover a range typical for aerosol size distributions often encountered in the summertime high Arctic (Heintzenberg and Leck, 2012; Leck and Svensson, 2015). If we assume it is considered that the accumulation mode contributes most to atmospheric CCN, then the simulation with 20 cm^{-3} of both accumulation and Aitken mode aerosols agrees the best with the observations in terms of the CCN concentrations measured onboard the ship (Sect. 2.2). In this study, the simulation AC20_AK20 is ~~thus defined~~ considered as the baseline simulation.

A full description of meteorological conditions during the ASCOS campaign is available in Tjernström et al. (2012). ~~During the ASCOS ice-drift period, observations were done over a surface dominated by pack ice and the surface conditions are thus set to represent sea ice in both models.~~ The values used for the surface temperature and surface pressure are 269.8 K and 1026.3 hPa, respectively. ~~The surface albedo is set to 0.844.~~ The observed turbulent fluxes were usually smaller than 5 W m^{-2} with peaks in the probability distributions around zero. Accordingly, the ~~thus prescribed~~ sensible and latent heat fluxes are set to zero (cf. also Stevens et al., 2018) ~~prescribed to zero.~~ ~~The surface roughness is set to 0.0004 and the surface albedo to~~ ~~albedo is set to 0.844~~ (cf. Sedlar et al., 2011). In both models, ~~Over the whole model domain,~~ the large-scale divergence is set to $1.5 \times 10^{-6} \text{ s}^{-1}$ at all model levels, which is the value required to obtain a stable cloud layer in both models (cf. Stevens et al., 2018). ~~There is no large-scale advection in the models.~~ The aerosol population in both modes is represented by lognormal functions with the distribution parameters based on the ASCOS campaign measurements (Igel et al., 2017). Modal diameters of 32 and 93 nm and standard deviations and 1.1 and 1.5 are used for the Aitken and accumulation modes, respectively. The simulations are initialized with prescribed cloud droplet number concentration equal to 30 cm^{-3} and the cloud water mixing ratios ~~ss~~ derived from the observations, i.e. a cloud is present at the beginning of all simulations. ~~All microphysical processes are active at the beginning of the~~

1110 simulations in both models. The prescribed ice crystal concentration is 0.2 L^{-1} (cf. Stevens et al., 2018). The model
1115 The 3D model domain is three-dimensional and covers a region of $6 \times 6 \times 1.7 \text{ km}^3$. In the horizontal direction,
there is a fixed grid distance of 62.5 m in both models. In the vertical direction, the grid in MIMICA is variable
with the highest resolution (7.5 m) at the surface and in the cloud layer. In RAMS, the vertical grid spacing is kept
constant at 10 m. The different default configurations of vertical grid spacing in the two models could potentially
generate discrepancies in the simulated cloud properties. However, we performed an additional baseline simulation
with a fixed vertical resolution of 10 m with MIMICA and found no significant differences in the simulated results
compared to the default version. The simulation period is 12 hours, where the first 2 h are assumed considered
to be a spin-up period and is therefore excluded from the figures and analysis. After the spin-up period, the cloud
layer is stable in the baseline simulations.
~~and they are therefore excluded from the figures and analyses.~~

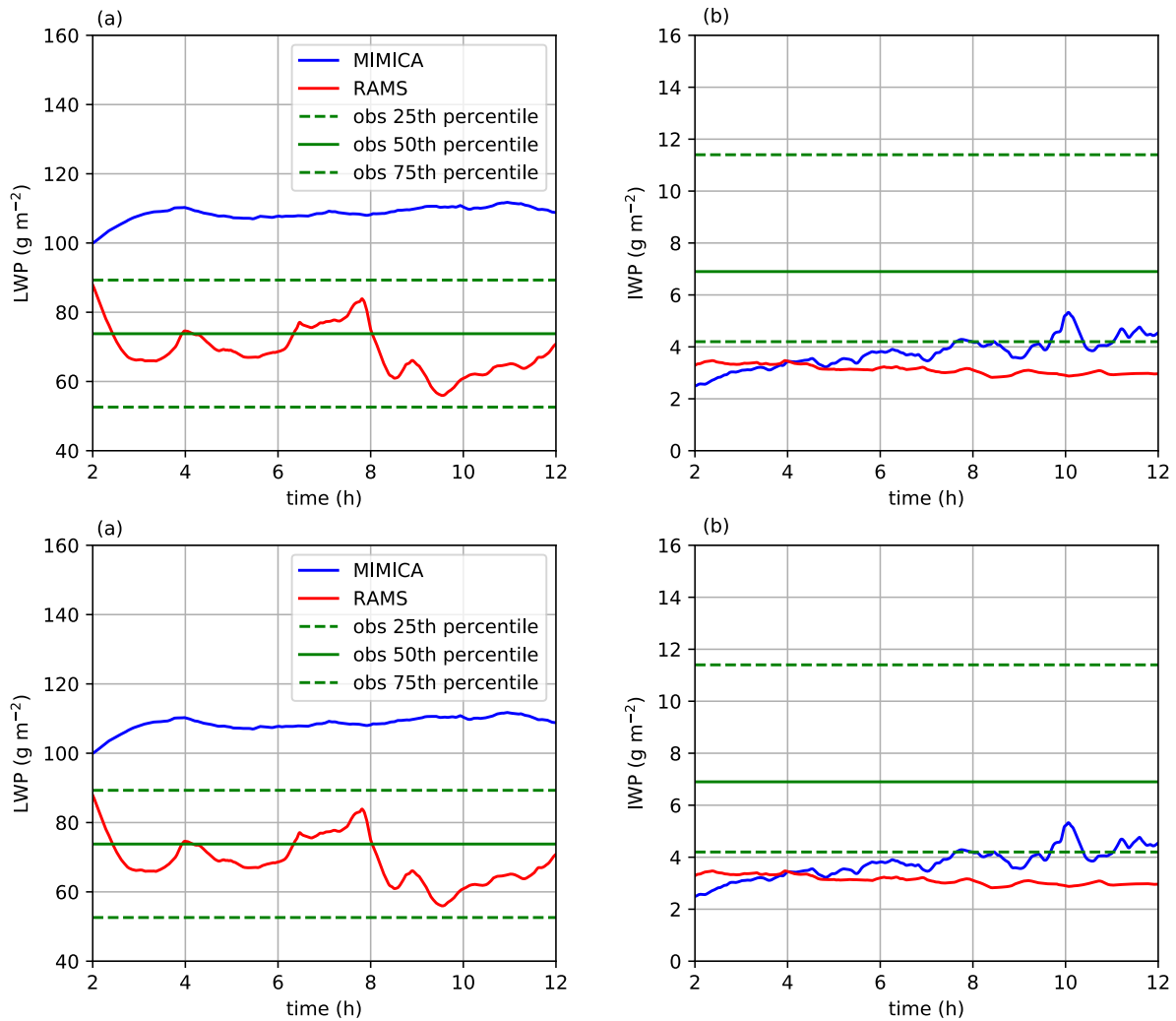
1120 –In clean environments, the source of Aitken mode aerosols is typically new particle formation (NPF) and
subsequent growth. In the high Arctic, however, different sources of Aitken mode aerosols have been proposed.
Some studies associate the Aitken mode with the NPF events and subsequent growth by DMS oxidation products
(e.g., Wiedensohler et al., 1996). Other studies suggest that the Aitken mode particles in this region are made up
of marine gels produced by phytoplankton and sea-ice algae at the sea-surface interface (Leck and Bigg, 2005b).
1125 Different mechanisms imply different chemical compositions and thereby different hygroscopic properties that
aerosol particles might have in the high Arctic. To study the impact of aerosol hygroscopicity, we have performed
additional simulations with different values of the hygroscopicity parameter, kappa, κ (Petters and Kreidenweis,
2007). The default κ -value used to describe the hygroscopic properties of both aerosol modes is set to 0.4 (Leck
and Svensson, 2015). As some previous studies (e.g., Christiansen et al., 2020) have shown that a change in
hygroscopicity of the accumulation mode aerosols has almost no influence on the cloud properties, we have only
1130 examined the sensitivity of the κ -value of the Aitken mode particles. Simulations AC3_AK20 and AC3_AK200
~~were have been~~ performed with two additional κ -values = [0.1, 1.1], which cover a typical range of hygroscopicity
of compounds expected to be present in high Arctic Aitken mode particles (Leck and Svensson, 2015). The lower
limit of the hygroscopicity parameter tested ($\kappa=0.1$) would be representative of e.g., many organic compounds
(e.g., Leck and Svensson, 2015) while the upper limit prescribed ($\kappa=1.1$) would correspond to a water-soluble
1135 inorganic salt like ammonium sulfate (Petters and Kreidenweis, 2007).

We have also ~~used MIMICA to~~ examined how the influence of Aitken mode particles on cloud microphysical
properties depends on the amount of ice present in the cloud. Additional versions of the simulations AC3_AK20
and AC3_AK200 ~~were have been~~ performed with prescribed values of the ice crystal concentrations set to 0 and 1
 L^{-1} . These values were chosen somewhat arbitrarily, but should generally represent a range describing an ~~relatively~~
1140 ice-free and an ice-rich cloud in the high Arctic. Note that the RAMS microphysical scheme includes hail and the
one in MIMICA does not. However, the riming process in RAMS is inefficient for the examined conditions and
pure ice crystals dominate the simulated ice water budget. The hail contribution to total surface precipitation is
also two orders of magnitude lower than the rain contribution in RAMS.

1145 3 Simulation results

3.1 Baseline simulation: comparison of simulated cloud properties

1150 We first compare our baseline simulations (AC20_AK20) with time series of observed LWP and IWP (Fig. 2+).

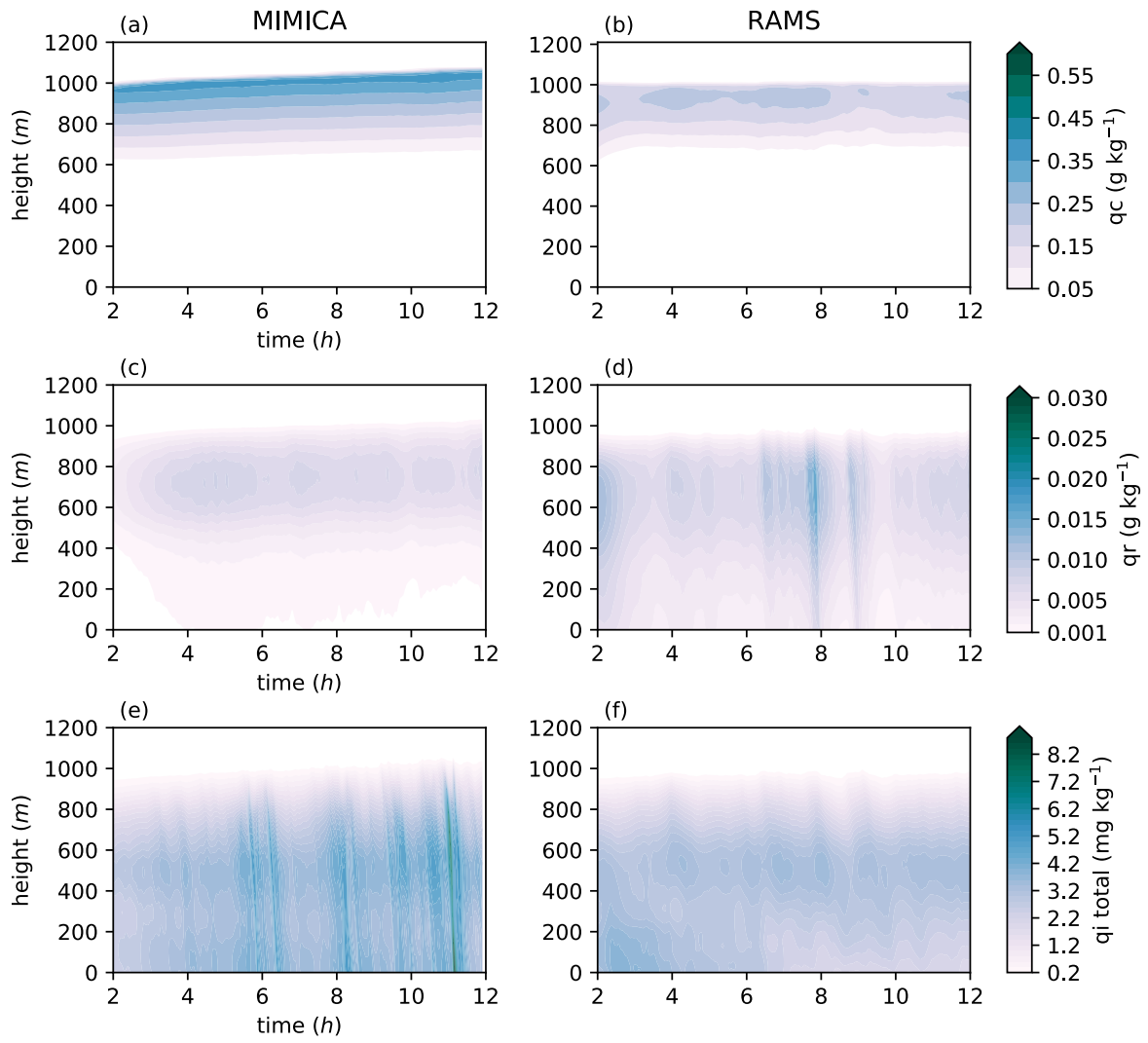


1155 **Figure 24:** (a) LWP and (b) IWP simulated by MIMICA and RAMS for the baseline simulations, i.e. with accumulation and Aitken mode concentrations of 20 cm^{-3} . The retrieved values of LWP and IWP for the observed **cloudy period defined in section 2.2 and up to the height of the model domains** are shown as 25th, 50th (median) and 75th percentiles. The first 2 h of simulations, **considered as spin-up**, are excluded **as they are considered as a spin-up period**.

1160 RAMS produces LWP values that fall within the observed range whereas MIMICA simulates a LWP that is 12–25 % higher than the 75th percentile of the observed range (Fig. 24a). In general, the use of prescribed aerosol particle concentrations should result in a higher LWP than if the simulations were performed with interactive aerosol particle concentrations (e.g., Stevens et al., 2018). It may be that MIMICA is more sensitive than RAMS in this regard. Furthermore, RAMS simulates weaker radiative cooling rates than MIMICA, which should produce a lower LWP in RAMS compared to MIMICA (see Sect. 3.2). **Another possible reason for the discrepancy in the simulated LWP between the two models could be the different vertical resolutions in MIMICA and RAMS, which in MIMICA is higher in the cloud layer (Sect. 2.1).** The simulated IWP in both models is close to the 25th percentile of the observed range (Fig. 24b). In MIMICA, the IWP overlaps with the 25th percentile value in the second half of the simulation while in RAMS it is 17–33 % lower than **the 25th percentile**. Overall, the results show that both models simulate reasonable LWP and IWP compared to the observational data, however, it is hard to conclude which model is better due to the large uncertainty and variability of the retrieved cloud variables (cf. **also** Sect. 2.2).

1175 Simulated cloud droplet, rain and total ice mixing ratios for the baseline simulation are shown for the two models in Fig. 32. The cloud droplet mixing ratio increases towards the top of the cloud in both models, but MIMICA produces slightly higher values in the upper part of the cloud layer (Fig. 32a and 32b), **consistent with as was also reflected in the higher LWP values (Fig. 24a).** There is also a difference in the cloud top height evolution, **which in MIMICA, the cloud top height increases with time whereas it remains constant in RAMS. Fig. 2e and 2d display Rain mixing ratios are similar for the two models (Fig. 3c and 3d), but RAMS produces that RAMS slightly produces slightly more rain below the cloud than MIMICA.** Both models **also** simulate similar values of

total ice mixing ratios, although MIMICA produces a few stronger vertical bands after 6 h of simulation (Fig. 32e and 32f). This type of pronounced bands is a result of strong collection rates of rain drops by graupel, which appear at different times due to different temporal distributions of updrafts and downdrafts (Figure A8). To better understand the cloud dynamics, we have also examined the cloud diagnostics (i.e. mass transfer rates between gas and condensed phases) for cloud droplet water, rainwater and ice crystals (Fig. 43). In MIMICA, cloud droplet water has the highest condensation rates at the top of the cloud whereas in RAMS they are homogeneously distributed within the cloud layer (Fig. 43a and 43b). The reason is most likely the higher entrainment rates at cloud top in MIMICA (not shown) that bring more water vapor into the cloud from the moist air that is ~~usually present across the humidity inversion above, which caps the cloud-topped boundary layer~~ ~~the stratocumulus topped boundary layers in the high Arctic (Figure 1c Sedlar et al., 2012; Shupe et al., 2013).~~ Higher entrainment rates are also consistent with the higher cloud top cooling rates present in MIMICA. Below cloud base, there is first a thin layer of cloud droplet evaporation ~~present in both models.~~ In RAMS, there is also below which is a sub-cloud condensation layer ~~veryer in RAMS, which is.~~ This condensation layer is produced by weak sub-cloud convection (not shown). Even though the condensation in this layer is infrequent, the associated mean rates are of the same order of magnitude as the condensation rates within the main cloud layer. The pockets of condensation and evaporation present in the main cloud layer are well-correlated with updrafts and downdrafts and they tend to cancel each other in the mean (not shown). This is why the average condensation rate in the main cloud is of the same order of magnitude as the one in the sub-cloud layer in RAMS. However, if we consider the domain median instead of the mean, then the condensation rates are higher within the main cloud layer and they are zero below the evaporation layer also in RAMS ~~in RAMS~~ (not shown). In both models, the condensational growth of raindrops is limited to the upper part of the cloud layer, while the maximum evaporation rates are found around the cloud base (Fig. 43c and 43d). This typically happens when the environment is subsaturated for liquid water but supersaturated for ice, which results in evaporation of raindrops and ~~the~~ growth of ice crystals. Ice crystals grow throughout the whole cloud layer with the highest deposition rates around cloud base (Fig. 43e and 43f), which corresponds well to the highest rain evaporation rates that are clearly seen ~~in the MIMICA results~~ (Fig. 43c). The ice crystal deposition and sublimation rates are higher in RAMS than in MIMICA since the two models partition the total ice deposition differently among ice hydrometeor categories (not shown). ~~Examining the total ice deposition/sublimation rates would most likely lead to similar rates between the two models, but these rates are not available in MIMICA.~~ Overall, ~~t~~he comparison shows that the simulated cloud microphysical properties are ~~all~~ within the same order of magnitude for the two models, i.e. both models ~~in general~~ simulate the same cloud mechanisms leading to cloud dynamics that are similar in many aspects. However, there are still some notable differences that arise from the different model configuration descriptions.



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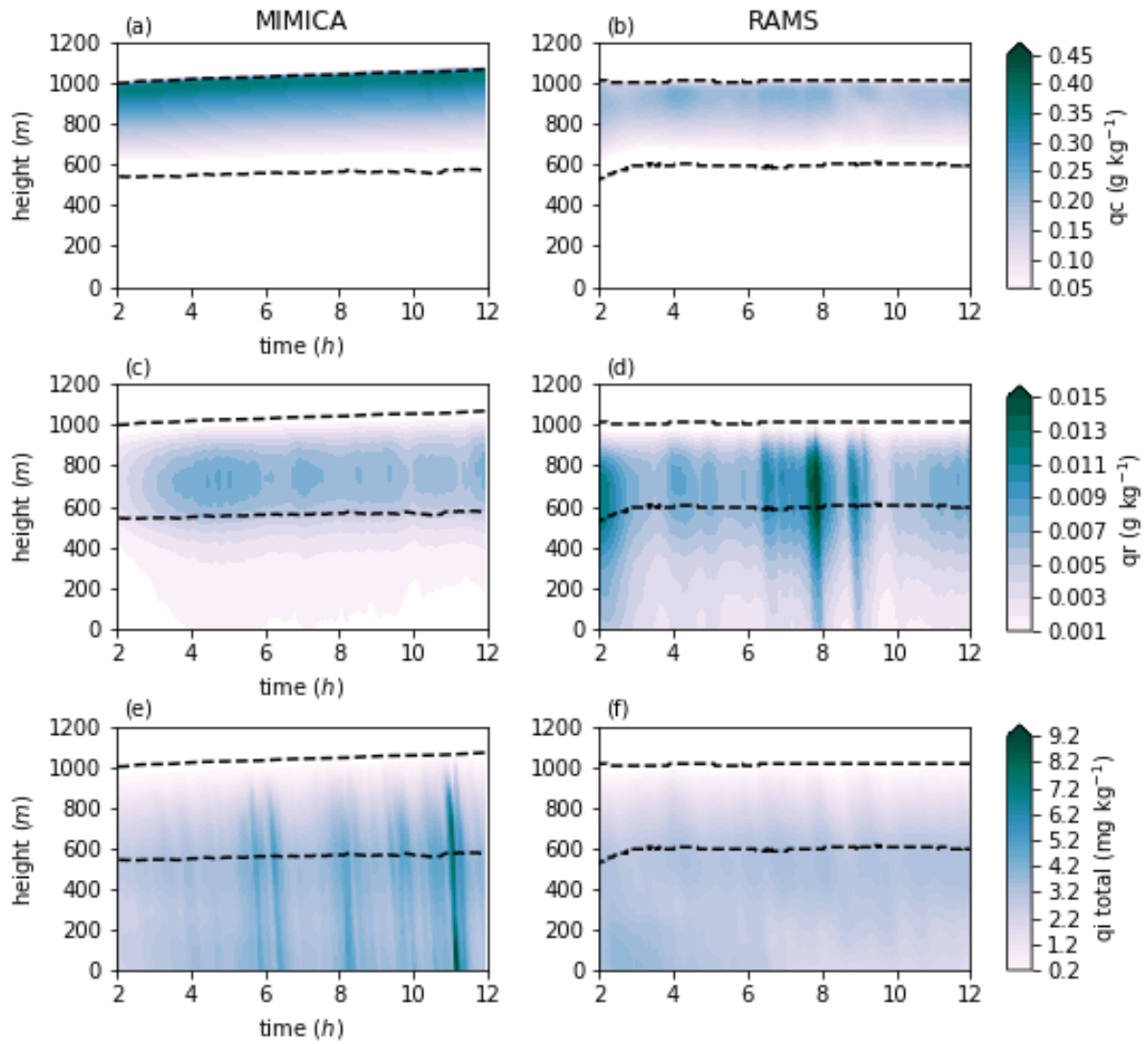
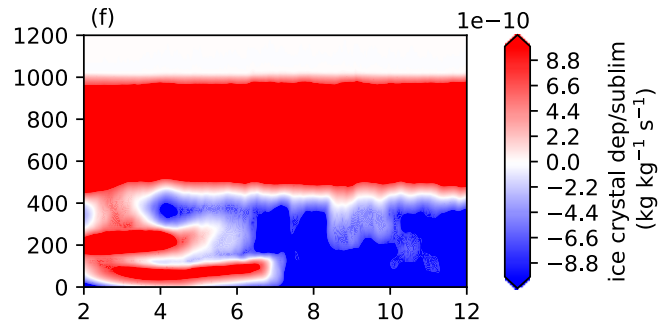
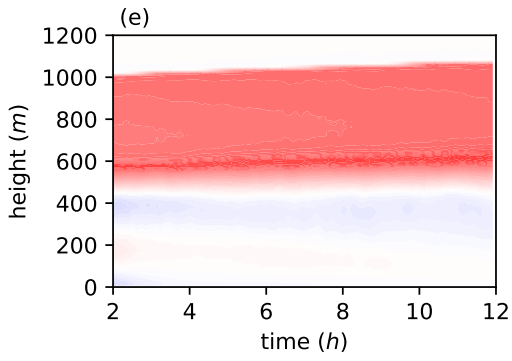
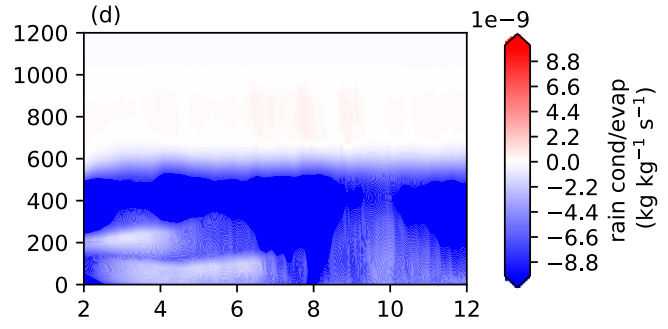
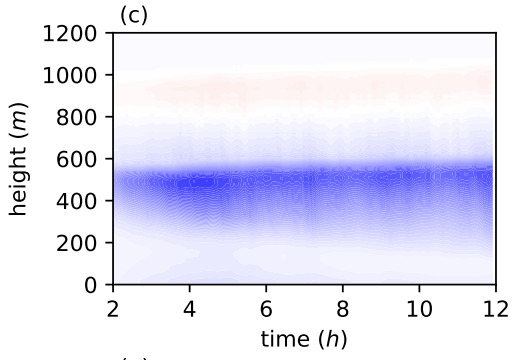
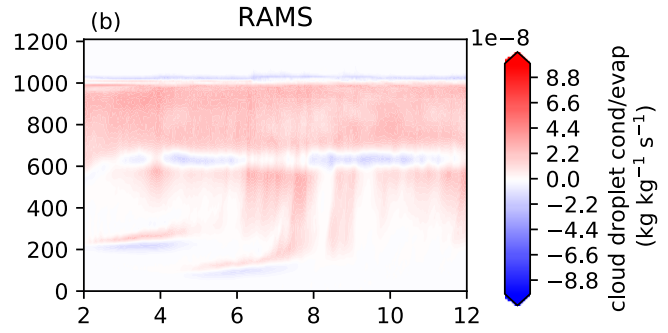
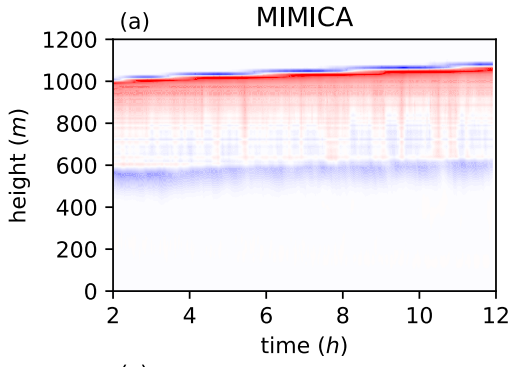


Figure 32: Cloud properties simulated by MIMICA and RAMS for the baseline simulation, i.e. with accumulation and Aitken mode concentrations of 20 cm^{-3} . (a,b) cloud droplet mixing ratio (q_c); (c,d) rain mixing ratio (q_r); (e,f) total ice (ice crystals, graupel (and hail for RAMS)) mixing ratio (q_i total). The first 2 h of simulations, considered as spin-up, are excluded, as they are considered as a spin-up period. Black dashed lines represent the cloud top and cloud base heights.



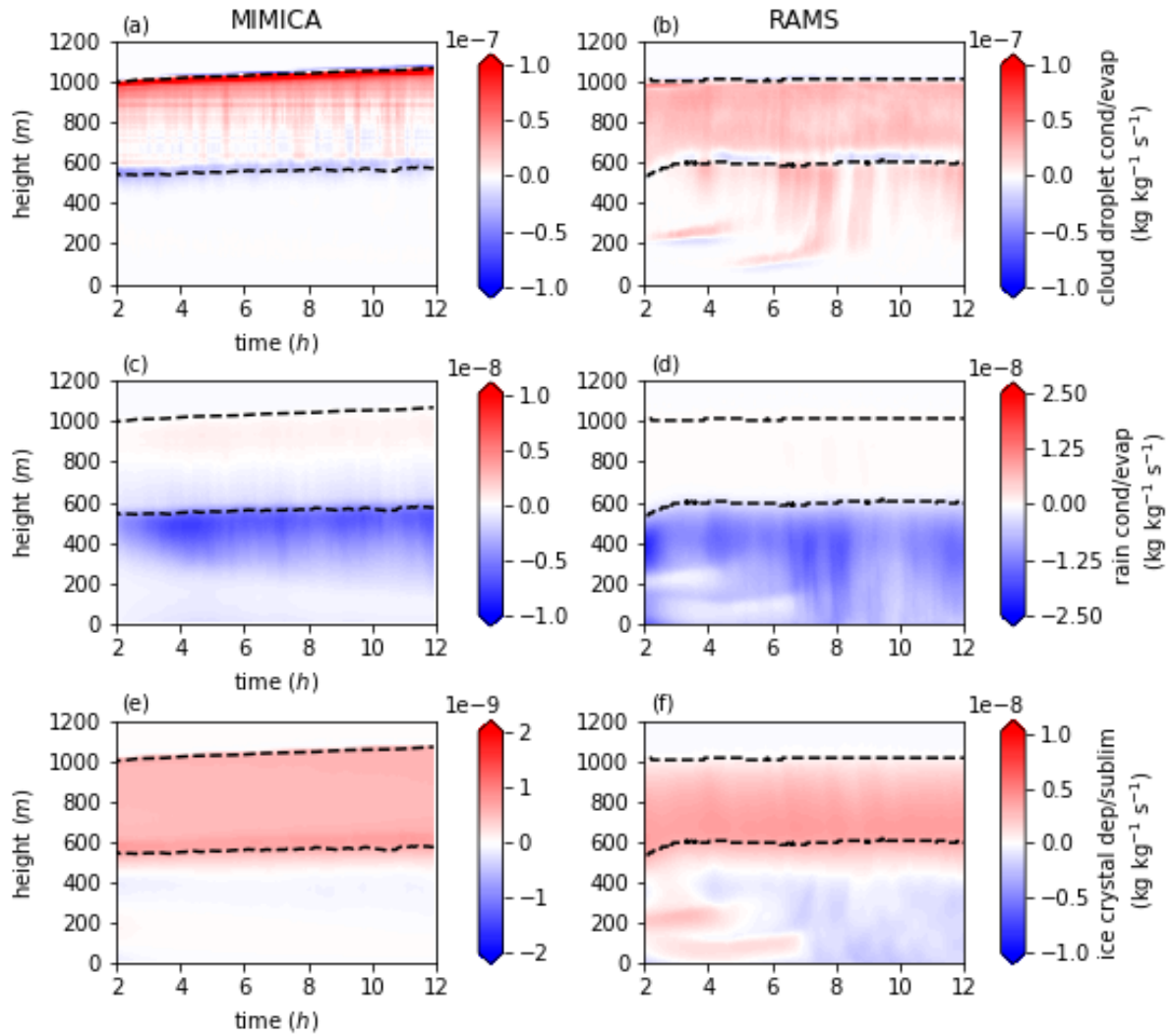


Figure 43: Cloud diagnostics simulated by MIMICA and RAMS for the baseline simulation, i.e. with accumulation and Aitken mode concentrations of 20 cm^{-3} . (a,b) cloud droplet condensation/evaporation rates; (c,d) rain drop condensation/evaporation rates; (e,f) ice crystal deposition/sublimation rates. The first 2 h of simulations, considered as spin-up, are excluded, as they are considered as a spin-up period. Red color indicates net condensation/deposition and blue net evaporation/sublimation. Black dashed lines represent the cloud top and cloud base heights.

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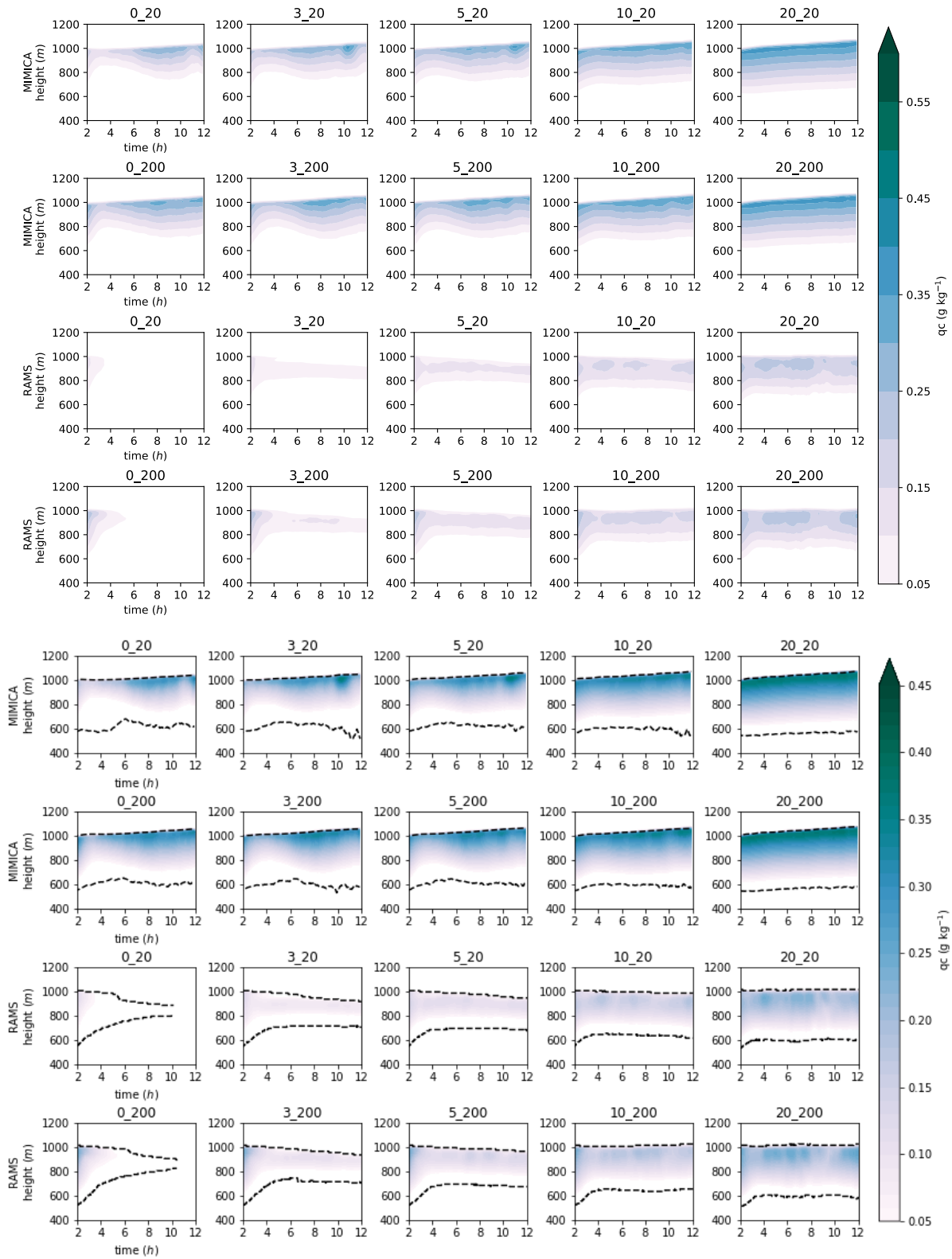
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3.2 Processes maintaining the simulated high Arctic SMP cloud

The cloud droplet mixing ratio for all simulations is shown in Fig. 54. In both models, the cloud thickens and the cloud base altitude change less with time when the number of accumulation mode aerosols increases. In general, MIMICA simulates a thicker cloud than RAMS. In both models, the cloud ~~thins~~shrinks at the beginning of the simulation (especially pronounced in the cases with low accumulation mode particle concentrations). However, the turbulence (Figure A6) is strong enough in MIMICA ~~is strong enough~~ to develop and maintain a stable cloud during the whole simulation with time for all cases. ~~This while this~~ is not ~~the case~~true for RAMS where the, ~~where~~ the cloud dissipates ~~completely~~ in the ~~simulations~~ with 0 cm^{-3} of accumulation mode particles (i.e. ~~the~~ AC0_AK20 and AC0_AK200 simulations). As one of the main generators of cloud turbulence is radiative cooling at the top of the cloud, we have compared the cooling rates between the two models. ~~They~~ The cloud top cooling rates in MIMICA are about two to three times greater in MIMICA than ~~those~~ in RAMS (Fig. 65 and A2) and are more similar in MIMICA to values obtained from radiative transfer calculations based on observational data (Brooks et al., 2017). For the baseline case, both models simulate a relatively thick cloud ($\text{LWP} > 40 \text{ g m}^{-2}$), which indicates that the differences in the cloud top cooling rates between MIMICA and RAMS do not arise from the difference in simulated LWP (cf., Garrett and Zhao, 2006). Most of the difference between the cooling rates is due to the cloud top liquid water content being about twice as large in MIMICA (Fig. 4). ~~Moreover~~However, the cooling rates in RAMS are smaller already even at the beginning of the simulations when the liquid water contents are very similar in the two models, ~~so the difference in liquid water content (Fig. 1a) cannot be the only explanation.~~ ~~A plausible explanation could instead be a~~ less efficient radiative cooling parameterization in RAMS than in MIMICA. To further investigate the influence of the radiation parameterization on the model results, we performed additional simulations with simplified radiative transfer schemes in both MIMICA and RAMS as well as simulation with a prescribed higher cloud top cooling rate in RAMS (Appendix A, Fig. A1). ~~could explain the discrepancy in simulated cloud droplet mixing ratios since lower radiative cooling at the top of the cloud leads to less turbulence in the cloud layer and consequently to less cloud water (as the free troposphere in the simulated case is a source of moisture to the cloud layer, cf. Sect. 1).~~ To further investigate how the radiation parameterization can influence the model results, we have performed additional simulations with MIMICA and RAMS where the models were utilized with simplified radiative transfer schemes or with a prescribed higher cooling rate at the cloud top in RAMS (Appendix A, Fig. A1). These simulations show that the radiation parameterization significantly modifies the simulated liquid water content and could be the ~~an~~ cause of the observed ~~substantial~~ differences between the models.

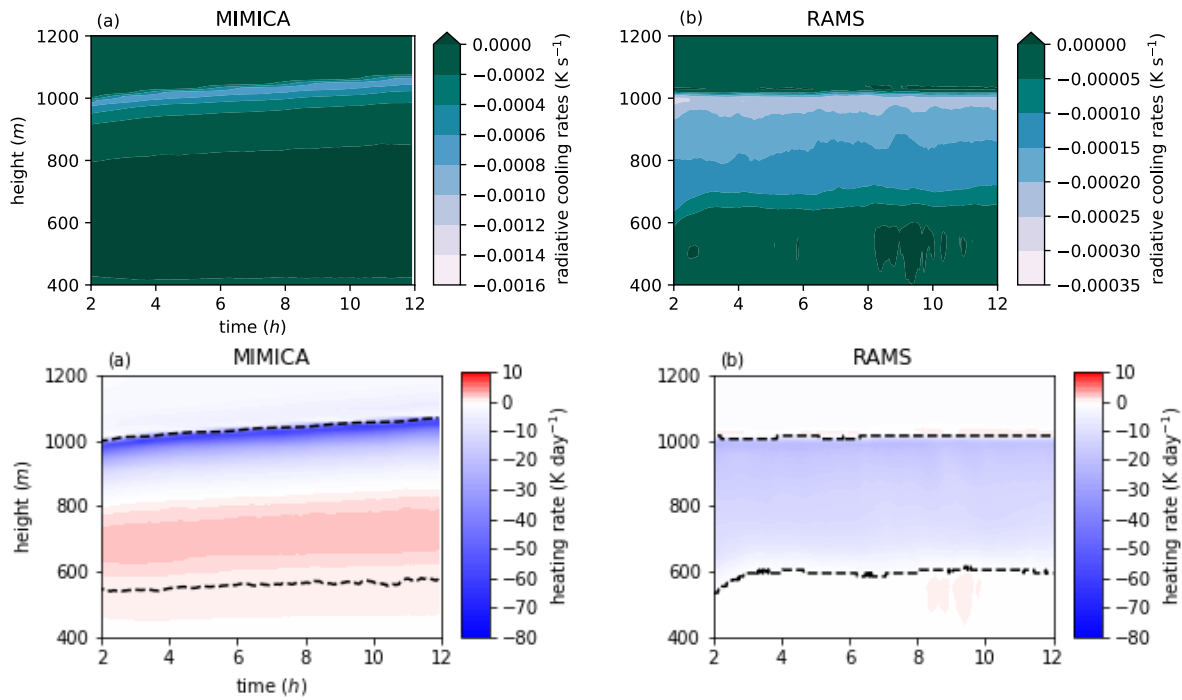
~~Figure 4 indicates a thicker cloud with a more stable cloud base when the number of accumulation mode aerosols increases. To clearly show the influence of Aitken mode particle concentration on simulated cloud microphysical properties, we have also shown the differences in cloud droplet, rain and total ice mixing ratios between each pair of simulations with the same accumulation mode concentration. These are discussed next (Sect. 3.3.1).~~



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Figure 54: Cloud droplet mixing ratio (q_c) for the MIMICA and RAMS simulation sets. The first 2 h of simulations, considered as spin-up, are excluded from the plots as they are considered as a spin-up period. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. “0_20” refers to “AC0_AK20”. **Black dashed lines represent the cloud top and cloud base heights.**

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1305 **Figure 65.** Radiative heating/cooling rates in the baseline simulation (AC20_AK20), simulated by (a) MIMICA and (b) RAMS. The first 2 h of simulations, considered as spin-up, are excluded as they are considered as a spin-up period. Black dashed lines represent the cloud top and cloud base heights.

1310 3.3 Influence of Aitken mode particles

1315 3.3.1 Influence of Aitken mode aerosol number concentration on cloud microphysical properties

1315 To clearly show the influence of the Aitken mode particles on simulated cloud microphysical properties we plot the differences in cloud droplet (Fig. 7), rain (Fig. 8) and total ice (Fig.9) mixing ratios between each pair of simulations with the same accumulation mode concentration. Figure 6 shows that adding Aitken mode particles generally increases the mass (Fig. 7) and number amount (not shown) of cloud droplet water generally increase in both models when Aitken mode particles are added, i.e. the particles serve as CCN and allow formation of additional cloud droplets. Aitken mode particles can also sustain the cloud at least for 6h when no accumulation mode particles present (Fig. 7). The extent of their influence depends on the concentration of accumulation mode particles since these particles activate more easily and have the primary control on the cloud droplet number concentration. A higher number of cloud droplets decreases the maximum supersaturation and the amount of available water vapor in the cloud available for activation of smaller particles in the cloud. In both models MIMICA, the influence of smaller particles on cloud droplet mixing ratio thus generally decreases monotonically with increasing accumulation mode concentration (Fig. 75), top row). In RAMS, the cloud is very thin (or even dissipates) at low accumulation mode concentrations. Thus, there is no clear trend in the impact of the Aitken particles until the accumulation mode concentration is about 10 cm^{-3} , when there is a stable cloud (cf. Fig. 4). At concentrations higher than 10 cm^{-3} of accumulation mode particles, there is a similar trend in RAMS as in MIMICA, i.e. the influence of the Aitken mode becomes less pronounced with increasing accumulation mode particle concentration (Fig. 5, bottom row). Nevertheless, the differences in cloud droplet mixing ratio are statistically significant differences in cloud droplet mixing ratio are statistically significant (according to a student t-test with a 95 % confidence level on the time averages in the cloud layer) for all pairs of different Aitken mode concentrations in both models, except for the MIMICA pair AC20_AK200 and AC20_AK20 (according to a student t-test with a 95 % confidence level on the time averages in the cloud layer). In other words, both models show that Aitken mode particles have a significant impact on the cloud droplet mixing ratio, at least up to 20 cm^{-3} of accumulation mode particles in RAMS and at least up to 10 cm^{-3} in MIMICA. At the cloud top, a distinct maximum difference occurs in both models as the largest influence occurs in the uppermost part of the cloud (both in MIMICA and RAMS) where most of the cloud droplet water is present (cf. Fig. 4). The maximum difference at the very top of the cloud is mainly a result of higher cloud top heights in the simulations with a higher Aitken mode concentration (Fig. A3).

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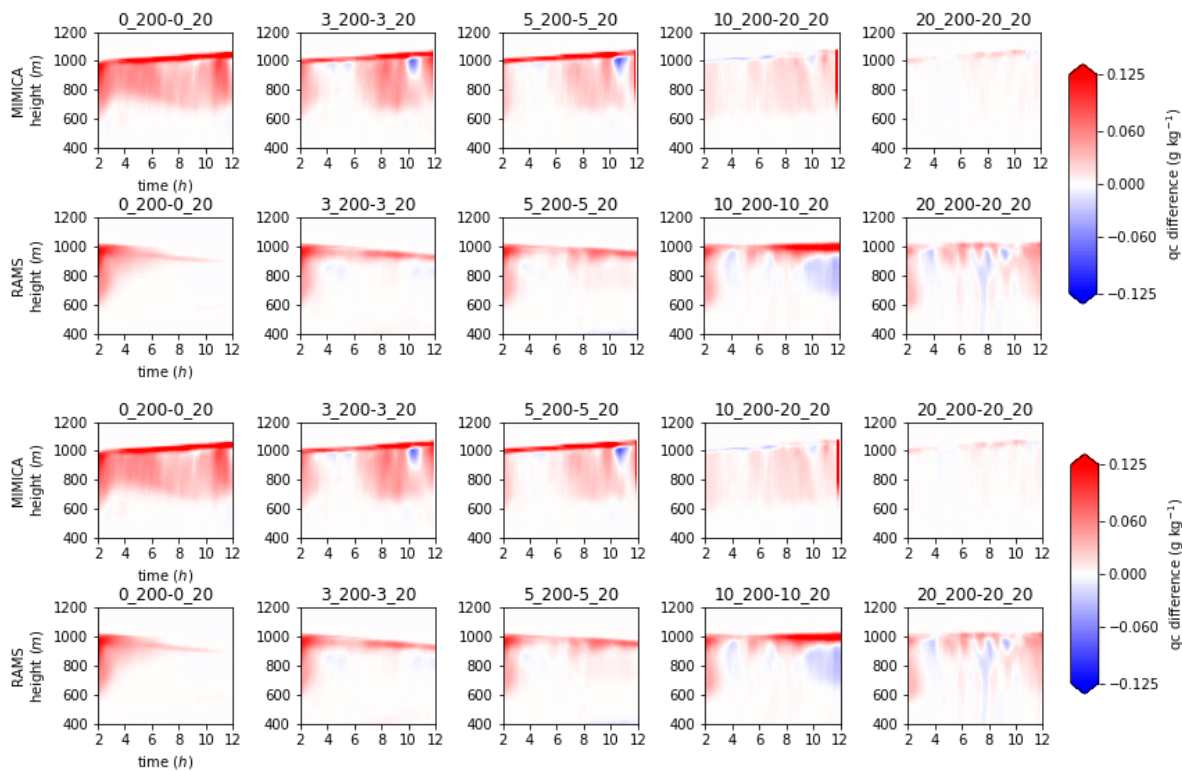


Figure 76: Differences in cloud droplet mixing ratio (q_c) for simulation pairs with the same accumulation mode concentration (i.e. $AC_x_AK200-AC_x_AK20$) shown for MIMICA and RAMS. The first 2 h of simulations, **considered as spin-up**, are excluded **as they are considered as a spin-up period**. A student's t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations, except for the pair $AC20_AK200$ and $AC20_AK20$ in MIMICA. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "0_20" refers to "AC0_AK20".

Most of the rain water is present in the upper part of the cloud layer in all simulations with MIMICA (Fig. A4), in line with the maximum rain condensation rates shown for the baseline simulation (Fig. 43). In RAMS, the cases with higher accumulation mode concentrations, i.e. with a stable cloud, also show that most of the rain water is present close to the cloud top (Fig. A4). Both models produce both positive and negative differences in rain water mixing ratio that vary with time with increasing Aitken mode particle concentrations (Fig. 8) Figure 7 shows the effect of Aitken mode particles on rain water, i.e. comparing the pairs of simulations with the same accumulation mode concentrations. There are both positive and negative differences that vary with time in both models. At the beginning of the simulations, there is in general more rain produced in the cases with a higher number of Aitken mode particles (i.e. positive differences). Despite the higher number of cloud droplets, An increase in the Aitken mode particle concentration may lead to stronger turbulence (Fig. A6) and more cloud liquid water production (Fig. 5), which could lead to stronger rain rates at the beginning of the simulations (cf. also Fig. A4). Towards the end of the simulations, the rain rates are either about the same or there is less rain (i.e. negative differences). The presence of both positive and negative differences with time is a result of differences in cloud dynamics with different temporal evolutions/distributions of updrafts and downdrafts with time in each individual simulation, which that influences govern the production of rain rain and ice production in the cloud (cf. Fig. A8b,c9).

The total ice mixing ratios for all simulations are presented in Fig. A5, while the differences in ice due to different Aitken mode concentrations are shown in Fig. 98 while total ice mixing ratios for all simulations are presented in Fig. A5. For the two lowest accumulation mode concentrations (0 and 3 cm^{-3}), there is in general more ice with a higher number of Aitken particles in both models. In the cases with more accumulation mode particles (e.g., 5, 10, 20 cm^{-3}), the variability is larger with both positive and negative differences. This result can again be related to differences in cloud dynamics; a change in the Aitken mode particle number concentration results in that maximum updrafts are reached at somewhat different times in the two model (cf. Figure A8b,c). The Differences are in general greater in MIMICA than in RAMS influence of Aitken mode particles on ice is in general larger in MIMICA than in RAMS consistent with the since there is a slightly more

total ice in MIMICA (Fig. A5), stronger cloud top cooling rates simulated by MIMICA that favors the ice formation through immersion freezing and growth by vapor deposition when the number of CCN increases (Possner et al. 2017; Solomon et al., 2018; Eirund et al., 2019). ~~but it is not possible to say which model is more realistic in terms of simulating the total ice amount (cf. Sect. 3.1).~~

The shown influence of Aitken mode particles on cloud microphysical properties and cloud sustenance has implications for the surface energy fluxes (Fig. 10). The influence of the smaller particles on the LW fluxes decreases as the number of accumulation mode particles increases (i.e. smaller differences) in both models, but it is statistically significant in all cases except for the pair AC20_AK200 and AC20_AK20 in MIMICA. The results are consistent with the influence of Aitken mode on cloud droplet mixing ratios (cf. Fig. 7). Both models simulate no significant influence of Aitken mode particles on the SW radiation at the top of the model domains, consistent with the low insolation (not shown).

We also tested the sensitivity of the simulated cloud properties to different Aitken mode particle concentrations for different levels of ice crystal concentrations—using MIMICA (see Sect. 2.3). These simulations show that ~~in clouds with more ice,~~ the influence of the Aitken mode particles on the liquid phase decreases in clouds with more ice becomes lower for the liquid phase (Fig. 11 and 12A6). This result agrees well with previous studies that have investigated the influence of CCN in mixed-phase clouds with different background INP or ice crystal concentrations (e.g., Possner et al., 2017; Stevens et al., 2018).

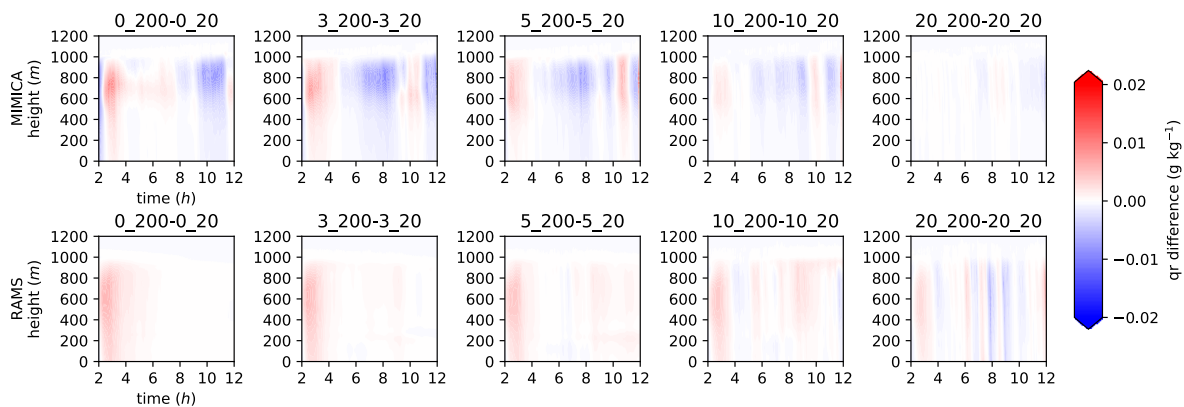
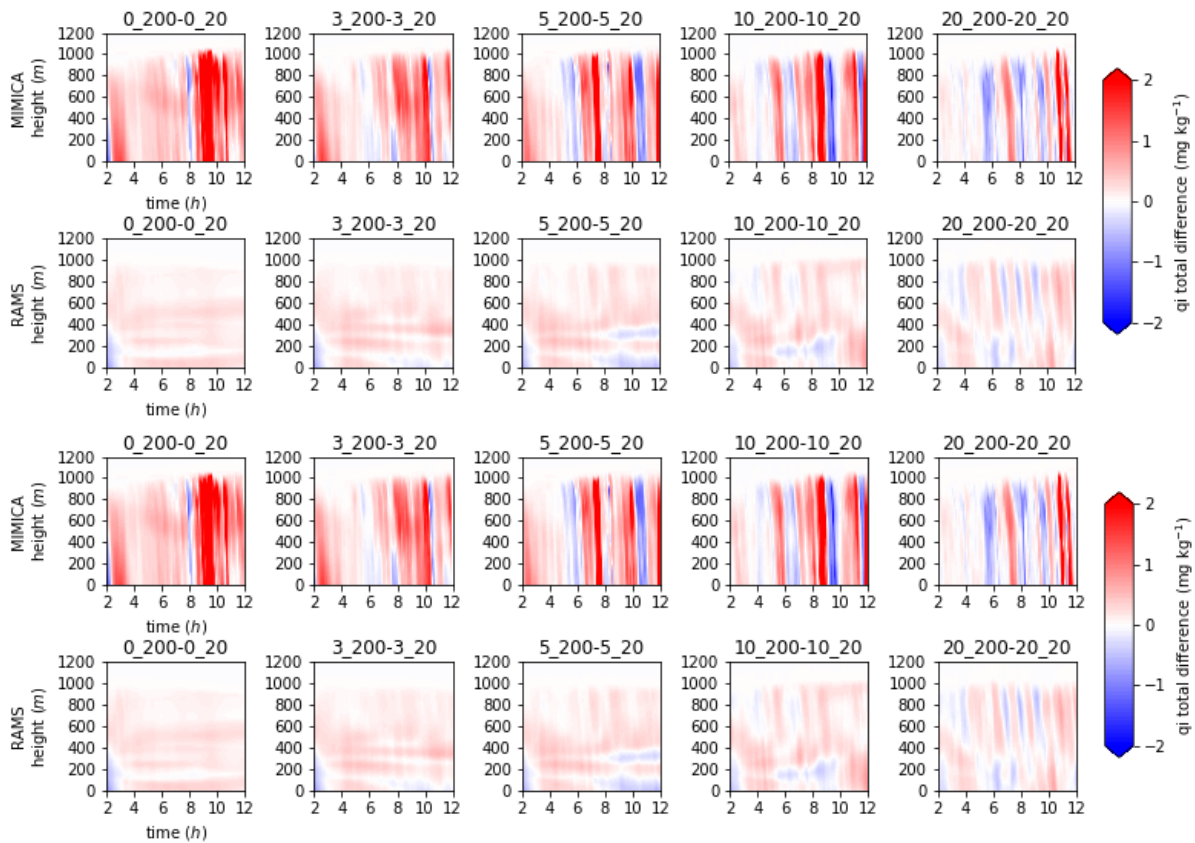


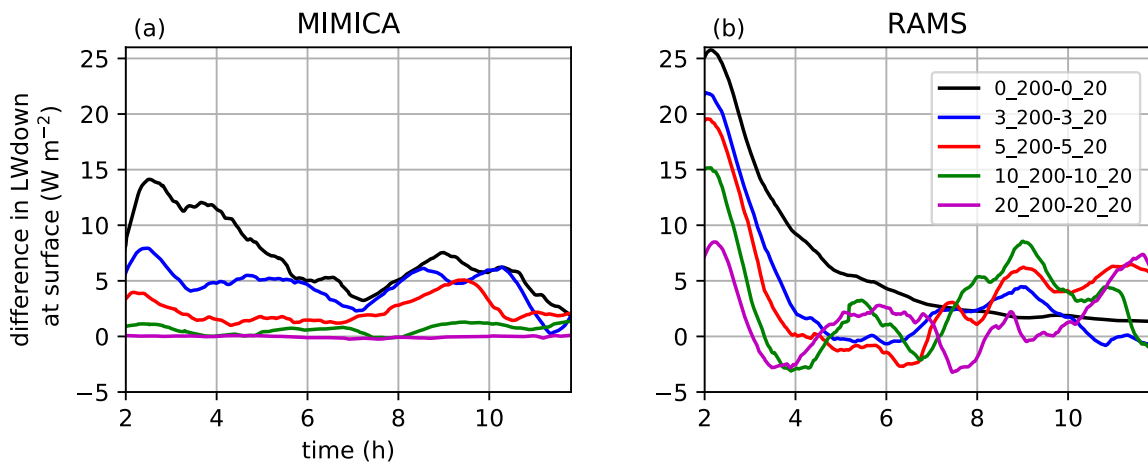
Figure 87: Differences in rain mixing ratio (qr) for simulation pairs with the same accumulation mode concentration (i.e. $AC_x_AK200-AC_x_AK20$) shown for MIMICA and RAMS. The first 2 h of simulations, ~~considered as spin-up,~~ are excluded ~~as they are considered as a spin-up period.~~ A student’s t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations except for the pair $AC20_AK200$ and $AC20_AK20$ in MIMICA. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. “0_20” refers to “ $AC0_AK20$ ”.



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Figure 98: Differences in total ice mixing ratio (q_i total) for simulation pairs with the same accumulation mode concentration (i.e. AC_x AK_{200} - AC_x AK_{20}) shown for MIMICA and RAMS. The first 2 h of simulations, **considered as spin-up**, are excluded **as they are considered as a spin-up period**. A student's t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different in the first four pairs of simulations in both models. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "0_20" refers to " AC_0 AK_{20} ".

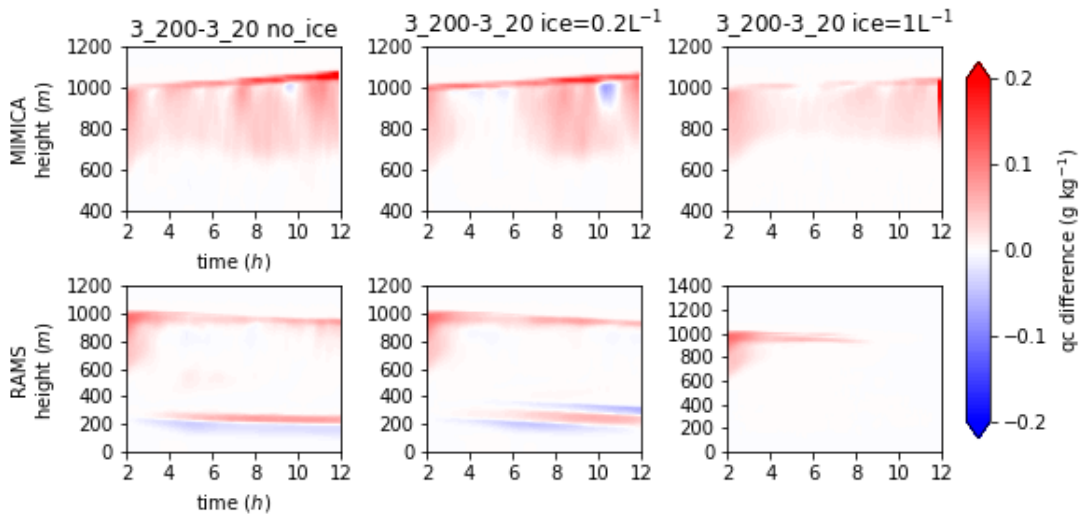
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Figure 10: Difference in downward longwave (LWdown) radiation at the surface for simulation pairs with the same accumulation mode concentration (i.e. AC_x AK_{200} - AC_x AK_{20}) shown for (a) MIMICA and (b) RAMS. The first 2 h of simulations, considered as spin-up, are excluded. A student's t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations except for the pair AC_{200} and AC_{20} AK_{20} in MIMICA. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "0_20" refers to " AC_0 AK_{20} ".

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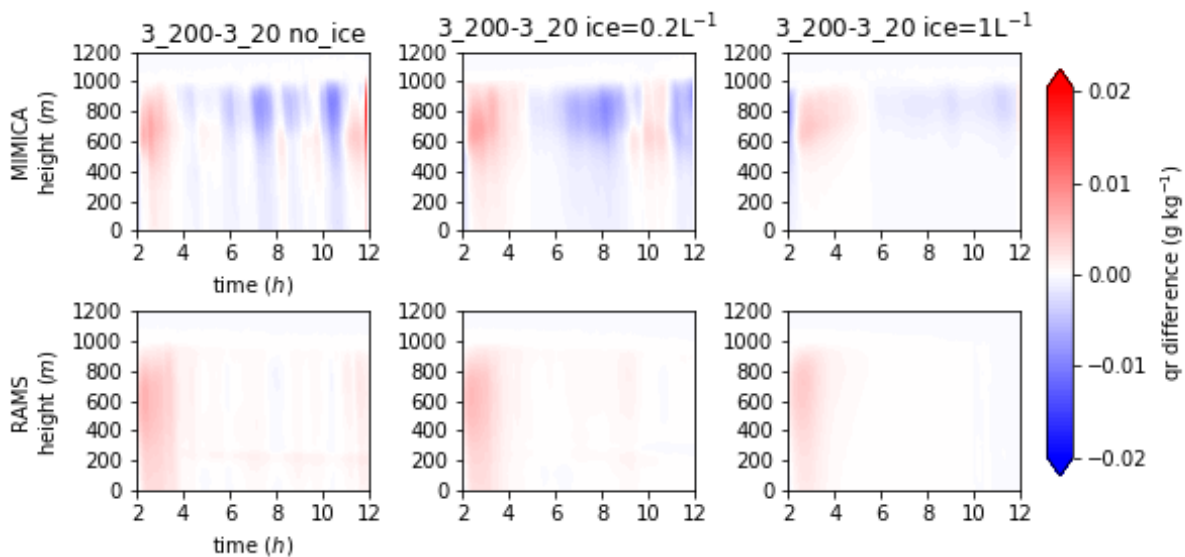


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Figure 11: Differences in cloud water mixing ratio (q_c) for simulation pairs with the same accumulation mode concentration and the same ice crystal concentration: $=0 \text{ L}^{-1}$ (no ice, the leftmost column); $=0.2 \text{ L}^{-1}$ (the middle column); $=1 \text{ L}^{-1}$ (the rightmost column) shown for MIMICA and RAMS. The first 2 h of simulations, considered as spin-up, are excluded. A student's t -test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "3 20" refers to "AC3 AK20".



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Figure 12: Differences in rain water mixing ratio (q_r) for simulation pairs with the same accumulation mode concentration and the same ice crystal concentration: $=0 \text{ L}^{-1}$ (no ice, the leftmost column); $=0.2 \text{ L}^{-1}$ (the middle column); $=1 \text{ L}^{-1}$ (the rightmost column) shown for MIMICA and RAMS. The first 2 h of simulations, considered as spin-up, are excluded. A student's t -test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations except for the pair with ice crystal concentration of 1 L^{-1} in RAMS. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "3 20" refers to "AC3 AK20".

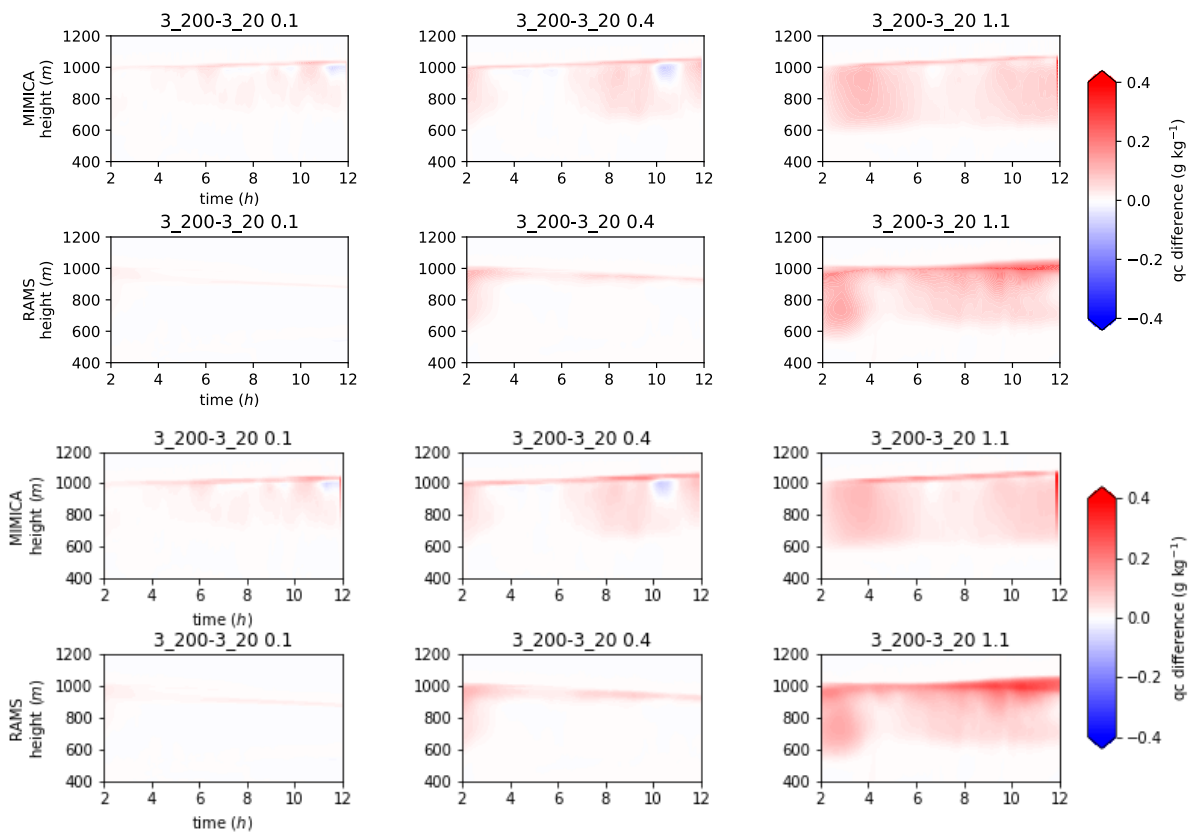
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3.3.2 Influence of Aitken mode aerosol hygroscopicity on cloud microphysical properties

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Figure 139 shows that the change in the cloud droplet mixing ratio induced by Aitken mode particles increases as their κ -value increases, i.e. more hygroscopic Aitken mode particles lead to a larger increase in the cloud droplet ~~mass amount~~. The cloud droplet number undergoes the same dependence (not shown). Higher particle hygroscopicity allows aerosol particle activation at lower supersaturations (see Sect. 4 for more information on supersaturation statistics).

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Figure 139: Differences in cloud droplet mixing ratio (q_c) for simulation pairs with the same accumulation mode concentration and the same kappa value of the Aitken mode particles: =0.1 (the leftmost column); =0.4 (the middle column); =1.1 (the rightmost column) shown for MIMICA and RAMS. The first 2 h of simulations, considered as spin-up, are excluded The first 2 h of simulations are excluded as they are considered as a spin-up period. A student's t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "0_20" refers to "AC0_AK20".

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The addition of Aitken mode particles with high κ -value leads to negative differences in rain amount in MIMICA, which can be explained by a greater number of cloud droplets and less efficient production of rain drops. However, in RAMS the differences are mostly positive, i.e. there is an increase in rain water mixing ratio amounts (Fig. A97). The reason for this is most likely probably the very weak cloud layer produced by RAMS in the original AC3_AK20 simulation. As there is no cloud, there is also almost no precipitation - regardless of the κ -value. In both models, the impact of Aitken mode particles on the total ice mixing ratio generally becomes greater as the hygroscopicity of the particles increases (Fig. A108).

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To summarize, the sensitivity tests show that Aitken mode particles can be activated even with a κ -value equal to 0.1 (more pronounced in MIMICA). Based on the model simulations, we can thus conclude that Aitken mode

particles do not have to be highly hygroscopic in order to become CCN-active if accumulation mode aerosol concentrations are low.

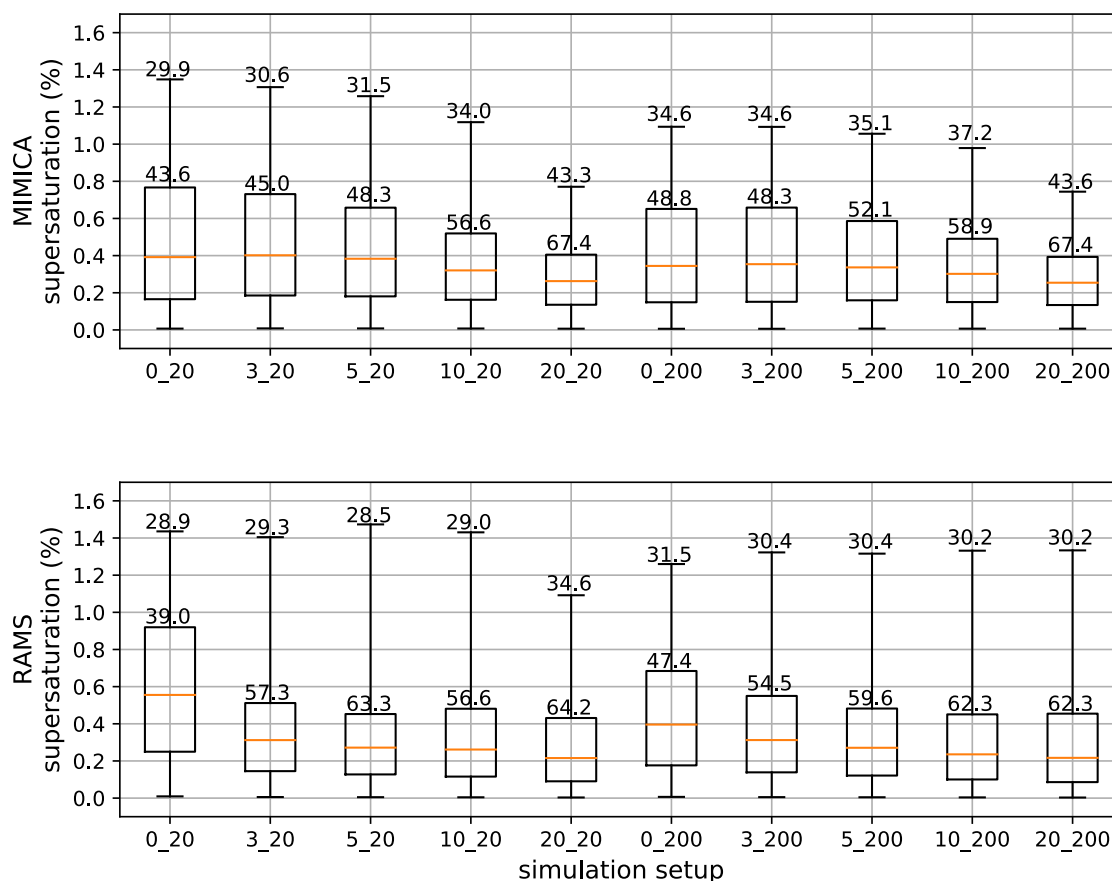
1495 4 Supersaturation statistics

1500 We analyze next the simulated water vapor supersaturation (ss) values reached within the model domains in order
1505 to investigate how the ss statistics depend on different prescribed aerosol size distributions as well as on different
hygroscopic properties of the Aitken mode particles. The ss statistics ~~archave been~~ calculated for a 20min period
around 6 h of simulation for all the cases simulated with the default κ -value ($\kappa=0.4$; i.e. dependence on the aerosol
size distribution, Fig. 140). They ~~are alsohave also been~~ calculated for the AC3_AK20 and AC3_AK200
simulations initialized with different κ -values for the Aitken mode particles ($\kappa=[0.1,1.1]$; i.e. dependence on the
hygroscopic properties, Fig. 154). In figure 140, the median ss values in both models generally vary between 0.2
and 0.4 %. The exception is the case AC0_AK20 in RAMS where there is no stable cloud at 6 h. The ss values in
this simulation are high since the statistics are based on a relatively low number of supersaturated grid boxes,
which in this case reach high ss values due to a low condensational sink. The median numbers agree well with
typical ss values reported for clean marine stratocumulus clouds at mid-latitudes (Hudson and Noble, 2014; Yang
et al., 2019). However, the 99th percentiles show high supersaturations with values above 1 % for most of the
simulations. As expected, simulated ss values decrease with higher accumulation mode number concentration.
1510 They are even lower when the Aitken mode concentration is prescribed to a larger number (200 vs. 20 cm⁻³). The
median values in Fig. 154 also vary between 0.2 and 0.4 % and in general decrease with a higher κ -value of the
Aitken mode particles for the two tested concentrations in both MIMICA and RAMS. Again, the 99th percentiles
show high values that exceed 1 % in most of the cases.

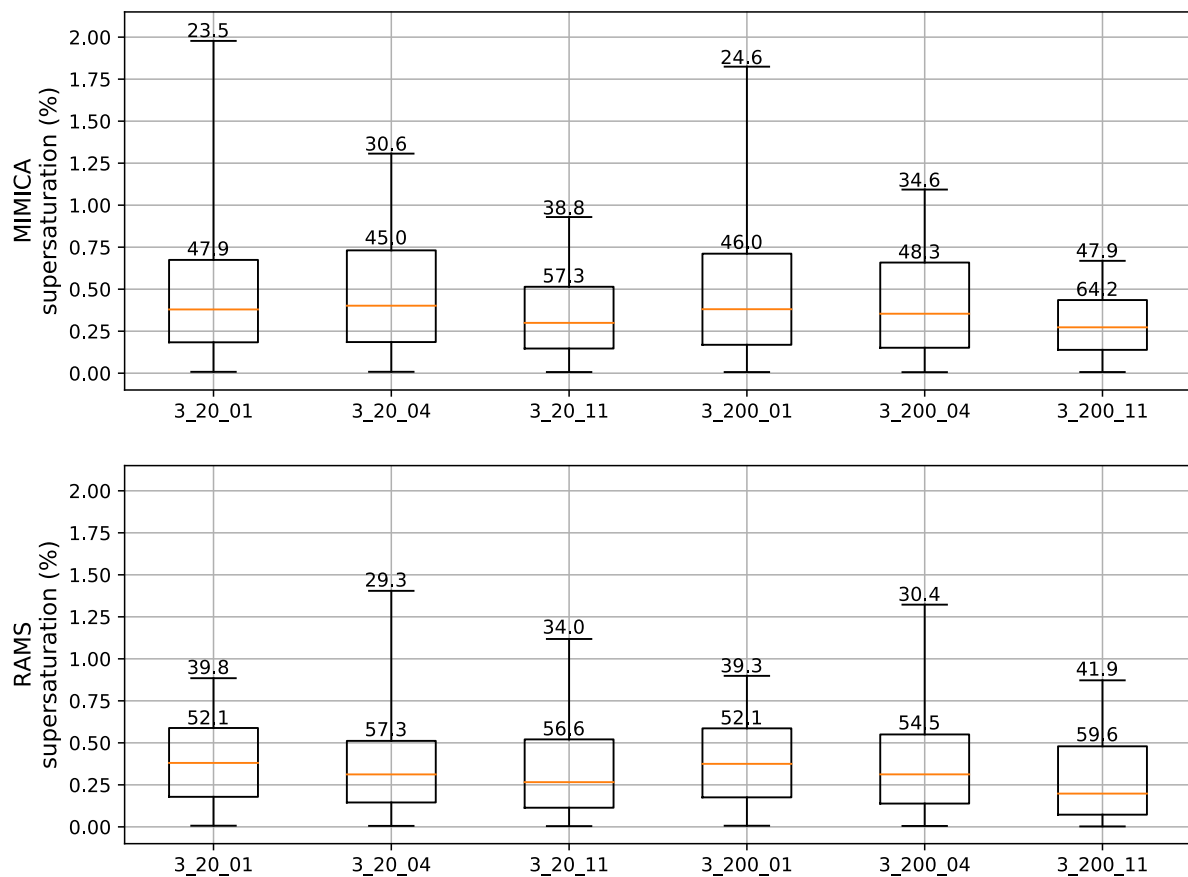
1515 The numbers shown in Fig. 140 and Fig. 154 are the critical dry diameters calculated for the 75th and 99th
percentiles of the ss values (cf. Fig. A10). These values can be ~~generally~~ compared with the mean diameter of 32
nm prescribed for the Aitken mode in this study. Our analysis confirms that supersaturations within the model
domains reach high enough values to activate Aitken mode particles for all tested accumulation mode
concentrations and all tested Aitken mode κ -values. ~~The results also agree well with recent findings for the region
south of the ice edge during the summertime, which have shown that particles smaller than the accumulation mode
potentially can act as CCN (i.e. smaller than 50 nm in diameter, Willis et al., 2016; Keeorius et al., 2019; Koike
et al., 2019).~~ If the calculations in Fig. A140 are done for higher (lower) surface tension, the maximum ss values
would need to be higher (lower) to activate particles of the same critical dry diameters as the ones presented here.

1520 Updraft statistics calculated for the same time period as the ss statistics for the set of cases simulated with the
default κ -value show that the updrafts are in general stronger with increasing accumulation mode concentration
1525 (Fig. A119). The ~~statistic~~ updraft values generally cover a range ~~of updrafts~~ between 0 to 1 ms⁻¹, which agrees
well with the vertically resolved updraft estimates by Sedlar and Shupe (2014) for the ASCOS campaign. With an
increase in accumulation mode particles, there is more vapor condensation and thus more liquid water in the ~~mixed-
phase cloud, which that~~ drives the turbulence through cloud-top radiative cooling (cf. Possner et al., 2017; Stevens
et al., 2018). Stronger turbulence further leads to stronger updrafts and further condensation.

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1535 | **Figure 140:** Supersaturation statistics shown for a set of cases initialized with a κ -value of 0.4, simulated by MIMICA and RAMS. The statistics are calculated for a 20min period around 6 h of simulation for all grid boxes with relative humidity > 100 %. Lower and upper whiskers correspond to 1st and 99th percentiles, respectively. The numbers written in the figure are critical dry diameters that correspond to supersaturation 75th percentiles (upper limit of the box) and 99th percentiles. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm⁻³, i.e. “0_20” refers to “AC0_AK20”.



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Figure 151. Supersaturation statistics shown for the simulations AC3_AK20 and AC3_AK200 initialized with different κ -values = [0.1, 0.4, 1.1], simulated by MIMICA and RAMS. The statistics are calculated for a 20min period around 6 h of simulation for all grid boxes with relative humidity > 100 %. Lower and upper whiskers correspond to 1st and 99th percentiles, respectively. The numbers written in the figure are critical dry diameters that correspond to supersaturation 75th percentiles (upper limit of the box) and 99th percentiles. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm⁻³, i.e. “3_20_01” refers to “AC3_AK20” initialized with a κ -value=0.1.

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1550 5 Discussion

1555 5.1 Qualitative comparison of model results with observational data for the High Arctic

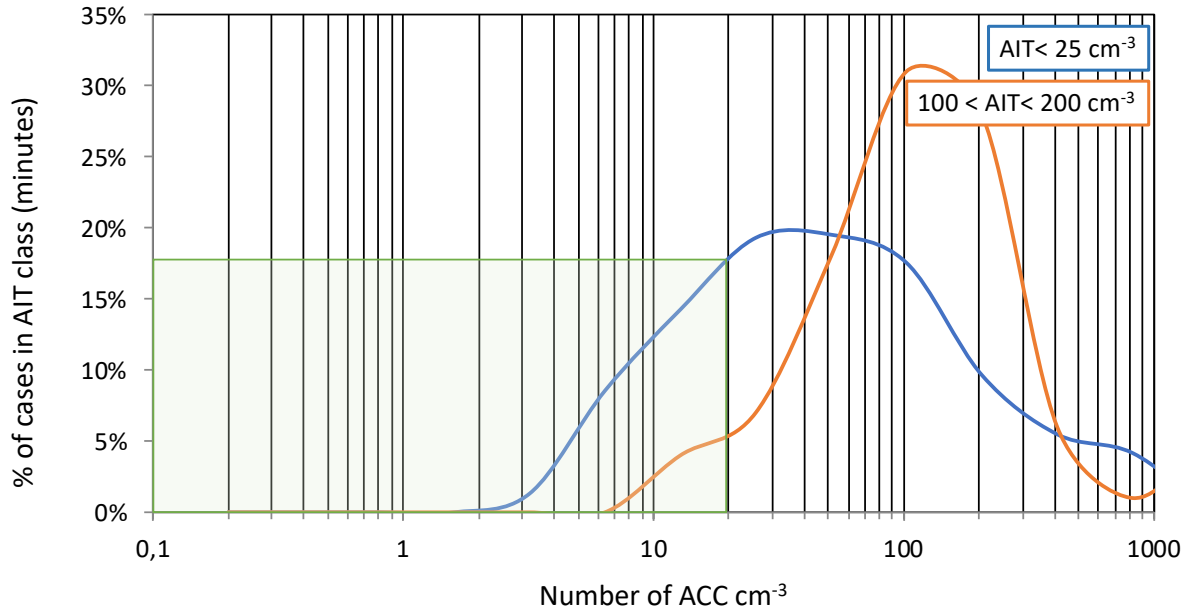
Both models suggest that Aitken mode particles are important as CCN in summertime high Arctic SMP clouds if accumulation mode concentrations are low. Guided by these analyses we have revisited the observed aerosol size distributions from four high Arctic expeditions, including the ASCOS campaign (Leck et al., 1996; Leck et al., 2001; Leck et al., 2004; Tjernstöm et al., 2014). We first examined the representativeness of the size distributions that we have applied in our simulations, i.e. how frequently these types of distributions occur in the observations. Fig. 162 shows two classes of size distributions: one with Aitken mode concentrations lower than 25 cm⁻³ (AIT < 25 cm⁻³, blue line) and one with Aitken mode concentrations between 100 < AIT < 200 cm⁻³ (orange line). The cases with accumulation mode number concentrations equal to 20 cm⁻³ (i.e. the maximum accumulation mode concentration prescribed in the simulations) have the occurrence probability of 5 % and 17 % (of total minutes of observations) for the class 100 < AIT < 200 cm⁻³ and AIT < 25 cm⁻³, respectively. This means that in conditions with low accumulation mode concentrations (i.e. lower than 20 cm⁻³) there is a higher probability for the Aitken mode particle concentration to also be low in number (i.e. lower than ~25 cm⁻³). However, it also happens that Aitken mode concentrations are much higher (>~100 cm⁻³). In other words, the prescribed size distributions that we have applied in our simulations are reasonable.

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Probability density functions (PDFs) of observed Hoppel diameters (Hoppel et al., 1986; Fig. 173), calculated as detailed in Heintzenberg and Leck (2012), show that the PDFs for all four expeditions peak around 60 nm, i.e. this

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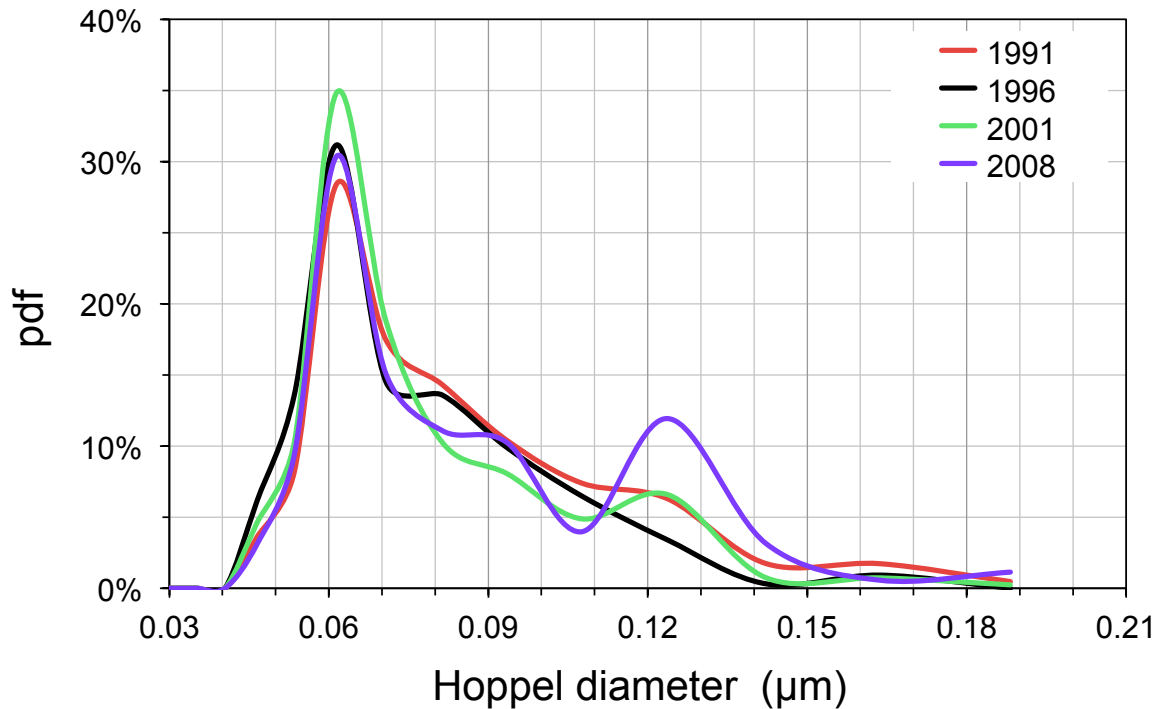
should be the most common activation diameter. However, the smallest observed Hoppel diameters are around 40 nm, supporting our conclusions that small Aitken mode particles may be activated in the summertime high Arctic under certain conditions. The observational statistics agree well with the calculations of the critical dry diameters obtained from the simulated ss values (Sect. 4).



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Figure 162. The occurrence probability (% of total minutes of observations) for two classes of Aitken (AIT) mode concentrations: $AIT < 25 \text{ cm}^{-3}$ and $100 < AIT < 200 \text{ cm}^{-3}$. On the x-axis is the number of accumulation (ACC) mode particles in cm^{-3} . The statistics are calculated for four different expeditions in the high Arctic, in the summers of 1991, 1996, 2001 and 2008. Further details on the quality and data processing of the aerosol size resolved measurements are available in Heintzenberg and Leck (2012).

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1585 **Figure 173:** Probability density function (pdf) of the Hoppel diameter shown for four different expeditions in the high Arctic, in the summers of 1991, 1996, 2001 and 2008. Further details on the quality and data processing of the aerosol size resolved measurements are available in Heintzenberg and Leck (2012).

1590 **5.2. General importance of Aitken mode particles for low-level mixed-phase cloud properties**

1595 Our study focuses on the summertime, ice-covered, high (> 80° N) Arctic region. However, it is reasonable to assume that the results are also valid for low-level mixed-phase clouds in other regions with low (<10-20 cm⁻³) accumulation mode aerosol concentrations. The activation diameters derived in Section 4 support recent findings for the region south of the ice edge during summertime, which show that particles smaller than the accumulation mode potentially can act as CCN (i.e. smaller than 50 nm in diameter; Willis et al., 2016; Kecorius et al., 2019; Koike et al., 2019). Several studies have investigated the seasonality of aerosol particle size distributions in the Arctic (e.g., Tunved et al., 2013; Freud et al., 2017; Koike et al., 2019). They show that number concentrations of accumulation mode particles are lowest during the summer and autumn months and that they can reach values below <10-20 cm⁻³ at several locations in the Arctic. However extremely low accumulation mode number concentrations (occasionally below 1 cm⁻³) have only been found in the high Arctic (Bigg et al., 1996; Mauritsen et al., 2011; Heintzenberg et al., 2015; Leck and Svensson, 2015). During summertime, conditions in the Arctic are generally favourable for NPF (Tunved et al., 2013; Croft et al., 2016; Nguyen et al., 2016) and local marine sources are active (Leck and Bigg, 2005a; Heintzenberg and Leck, 2012; Karl et al., 2013; Heintzenberg et al., 2015), supporting the presence of high concentrations of small particles. It is thus likely that Aitken mode particles are most important during the summer months (high concentrations of Aitken mode aerosols) and over remote areas covered by ice or snow (low accumulation mode aerosol concentrations).

1600 Note that the simulated influence of Aitken mode particles can also be dependent on details in the simulation setup. In this study, there are no sources or sinks of aerosols during the simulation time, the aerosols are only passively advected within the model domains (cf. Sect. 2.3). If aerosol sinks were included, the influence of Aitken mode particles would most likely be even more pronounced since accumulation mode aerosols are more efficient as CCN and should be removed faster from the cloud than the Aitken mode particles. Furthermore, the dependence of the Aitken mode influence on the cloud ice amount is investigated here based on different, prescribed ice crystal concentrations in the simulations (cf. Sect. 2.3). The results most likely depend on whether the ice crystal concentrations are prognostic or prescribed and thus could be different if we used prognostic ice crystal concentrations.

1620 **6 Summary and conclusions**

1625 This study investigates the potential importance of Aitken mode particles in sustaining and affecting the properties of stratiform mixed-phase clouds in the summertime high Arctic. To perform such a task, we have used two LES models (MIMICA and RAMS) to simulate a a-summertime high Arctic SMP cloud observed during the ASCOS campaign (Tjernström et al., 2014) and initialized the models with different aerosol size distributions. Both models show that Aitken mode aerosols have a significant impact on the simulated cloud droplet mixing ratio-droplet amount, if the accumulation accumulation-mode number concentration is less than 10–20 cm⁻³. Simulations performed with different values of the hygroscopicity parameter κ indicate that more hygroscopic Aitken mode particles lead to a higher amount of cloud droplet waternumber-of-cloud droplets, as expected. Moreover, the simulations show that Aitken mode particles can act as CCN and influence the properties of SMP clouds even at the low κ -values ($=0.1$). If the ice fraction of the SMP cloud is high (i.e. ice-rich clouds), the influence of Aitken mode particles on the liquid phase decreases, corroborating the results by Possner et al. (2017) and Stevens et al. (2018).

1630 =
1635 –Both models are in qualitative agreement in terms of the influence of Aitken mode particles on cloud properties, even though the models show different results regarding e.g., the simulated amount of liquid water and the relative role of different microphysical processes governing the overall cloud properties, and the simulated amount of LWP. The most striking difference in modelled cloud properties between the two models appears to be caused by a the difference in the radiation schemes. RAMS produces less radiative cooling for a certain amount of cloud water compared to MIMICA and does not sustain a cloud at low accumulation mode aerosol concentrations (<3–10 cm⁻³). The radiative cooling rates produced by MIMICA agree better with the observation-based estimates by (Brooks et al., (2017)), but however, the observations are are in general not sufficient to constrain or rank the models in

terms of their performance. This would require additional observations (of e.g., cloud-top radiative cooling rates, updrafts, supersaturation values) and less uncertainty in the retrieved data (of e.g., LWP and IWP).

The simulated median supersaturations in both MIMICA and RAMS vary between 0.2 and 0.4 %, but values above 1 % were also found within the model domains (99th percentile values). The spatial variability in the simulated supersaturations and updrafts demonstrates the potential issue of applying constant supersaturation values for a grid box, or even a certain cloud type, within e.g., general circulation models. Calculations of threshold diameters of aerosol activation confirm that the simulated supersaturation values are high enough for Aitken mode particles to be activated (i.e. the activation diameter is as low as ~30 nm). Furthermore, statistics of the observed Hoppel minimum diameter from four different expeditions in the high Arctic (Heintzenberg and Leck, 2012) also suggest that aerosols in the Aitken mode are activated as CCN. Our results are in qualitative agreement ~~also agree well with~~ the recent studies for the lower Arctic, which indicate that have inferred the importance of particles smaller than 50 nm act as a potential CCN (Willis et al., 2016; Kecorius et al., 2019; Koike et al., 2019), and thus suggest that Aitken mode aerosols more generally influence mixed-phase cloud properties in environments with low accumulation mode aerosol concentration.

—Our findings highlight the importance of better understanding Aitken mode particle formation, chemical composition ~~properties~~ and emissions, in particular in pristine environments such as the high Arctic in summer. It is reasonable to assume that the influence of these particles can be significant in any environment and during other seasons when the accumulation mode particle concentrations are low. The results ~~also~~ show that accumulation mode particles should not be considered as the only potential CCN in models, as this may lead to e.g., too low background CCN concentrations and too high estimates of anthropogenic aerosol indirect effects.

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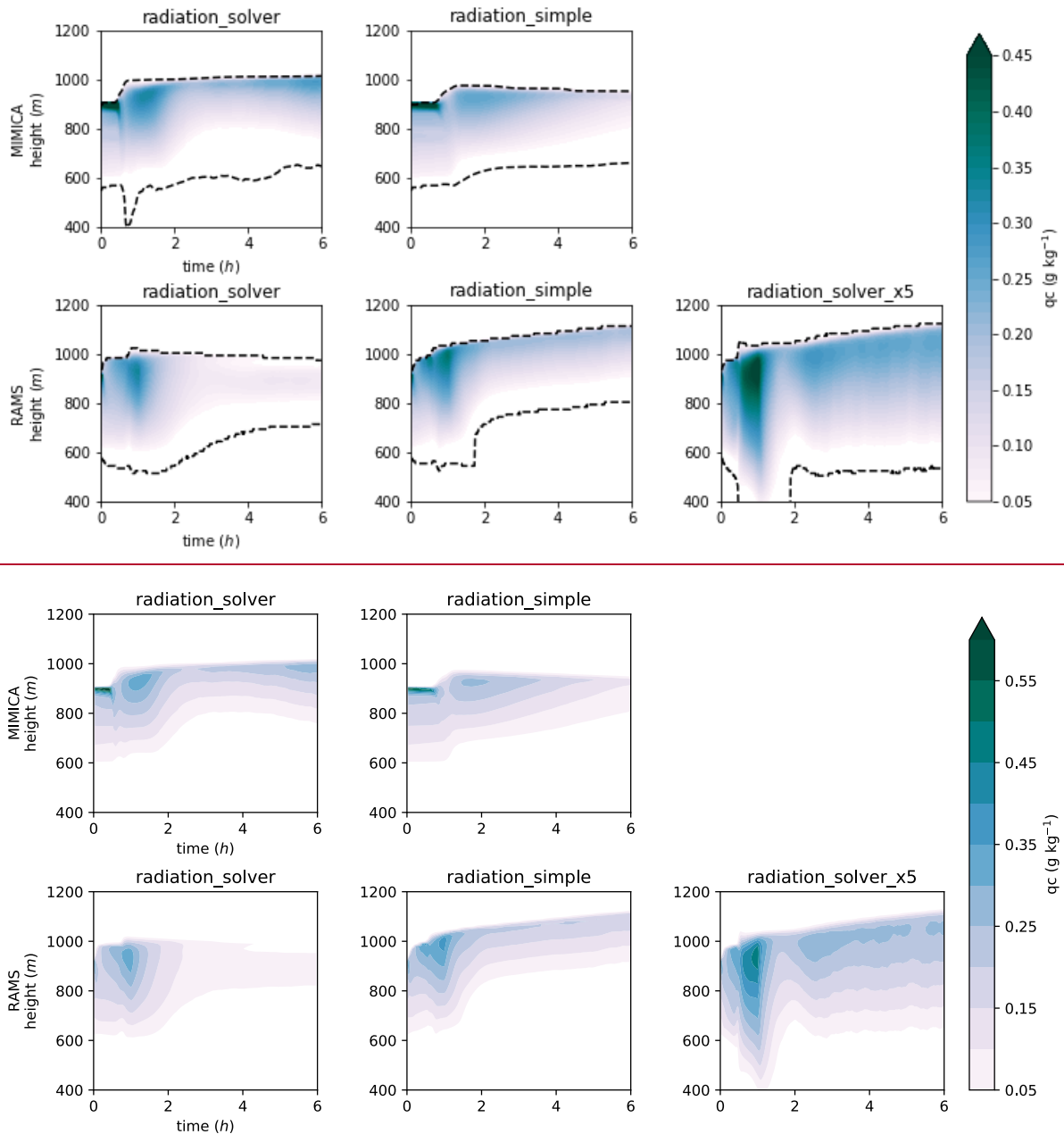
Appendix A:

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The cloud droplet mixing ratios simulated by the two models using different radiative transfer schemes is shown in Fig. A1. Using simple radiative transfer schemes (i.e. radiation_simple simulations; the radiative fluxes depend on LWP only, Stevens et al. (2005) in MIMICA and Chen and Cotton (1983) in RAMS) instead of the default radiation solvers (radiation_solver simulations; Fu and Liou (1993) in MIMICA and Harrington (1997) in RAMS) leads to a lower cloud water amount and a thinner cloud in MIMICA compared to RAMS, i.e. the opposite result compared to when using the default radiation solvers. Another test where the radiative cooling rates within RAMS were multiplied by a factor of 5 at the top of the cloud produces a much thicker cloud than the one in the MIMICA radiation_solver simulation, which confirms that the cooling efficiency of the radiative scheme is a critical factor for determining the cloud droplet amount and consequently also the cloud lifetime. The results show that the radiation parametrization used in the model has a significant impact on the simulated cloud properties and is especially important to be considered in model intercomparison studies.

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Figure A1: Cloud droplet mixing ratio (q_c) shown for a simulation AC3_AK20 initialized with different radiative schemes in MIMICA and RAMS. The title *radiation_solver* is used for the simulations where the models are initialized with their default radiation solvers (Fu and Liou (1993) in MIMICA and Harrington (1997) in RAMS). The title *radiation_simple* is used for the simulations where the radiative fluxes are calculated as functions of LWP only (Stevens et al. (2005) in MIMICA and Chen and Cotton (1983) in RAMS). The *radiation_solver_x5* simulated by RAMS shows the q_c obtained with the default radiation solver but with a 5x higher cooling rate enforced at cloud top. The simulations are run for 6 h. **Black dashed lines represent the cloud top and cloud base heights.**

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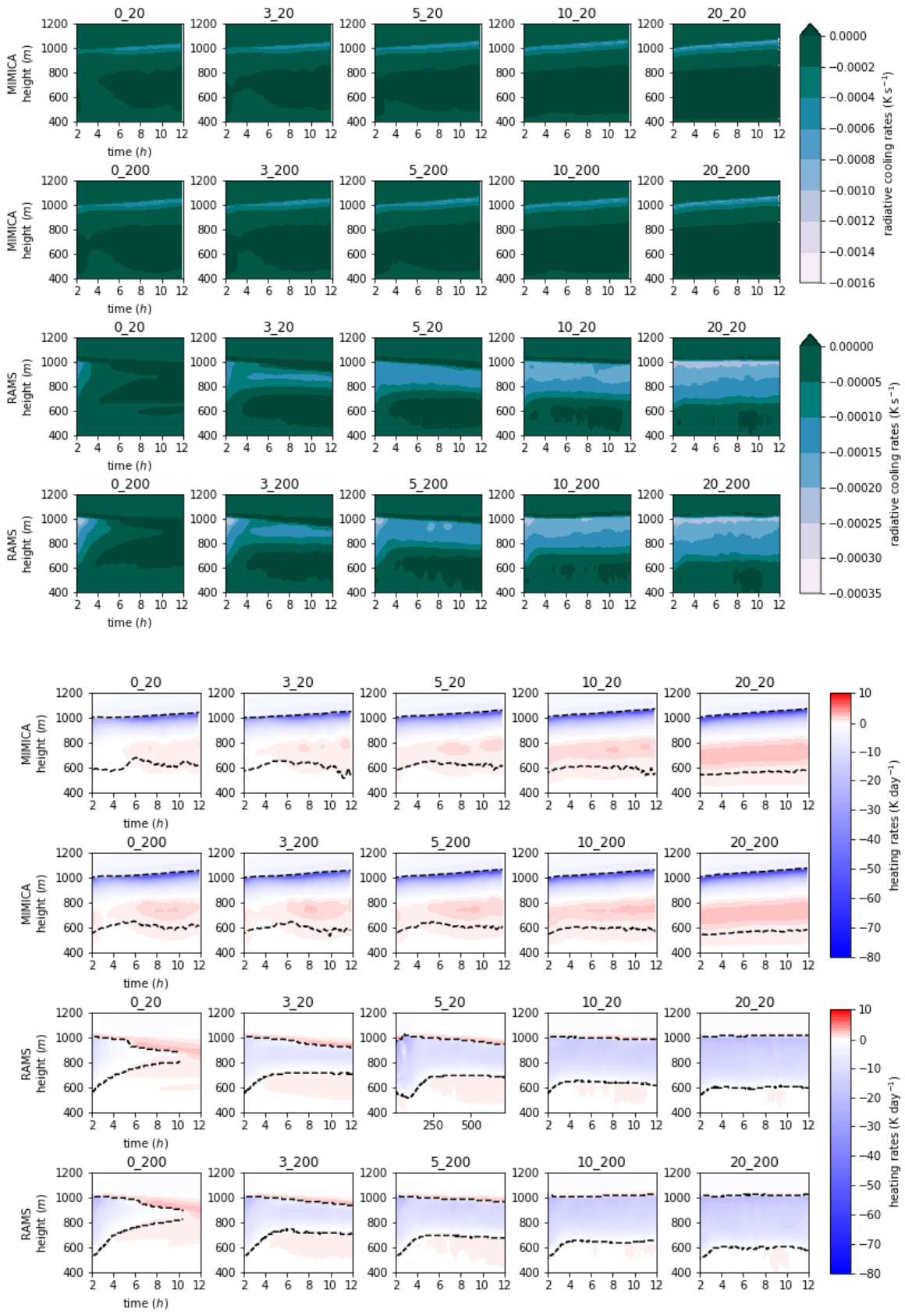
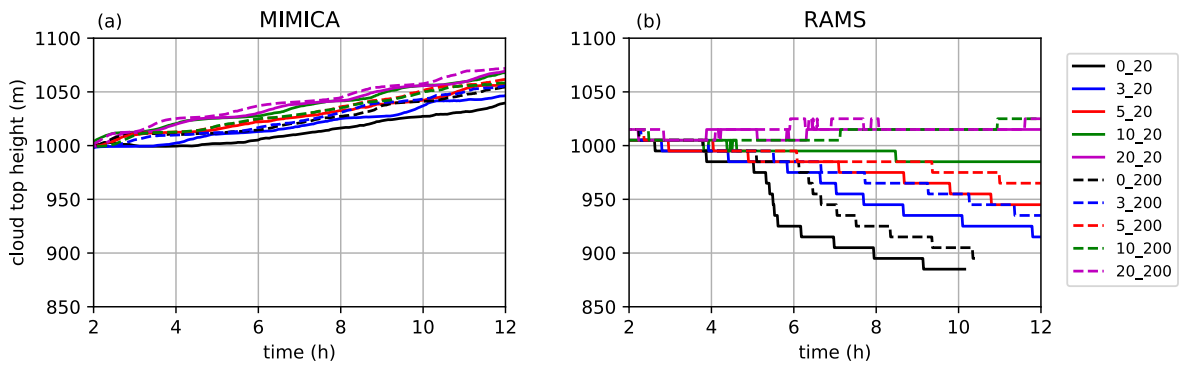


Figure A2: Radiative heating/cooling rates for the MIMICA and RAMS simulation sets. The first 2 h of simulations, considered as spin-up, are excluded from the plots as they are considered as a spin-up period. For figure clarity, the

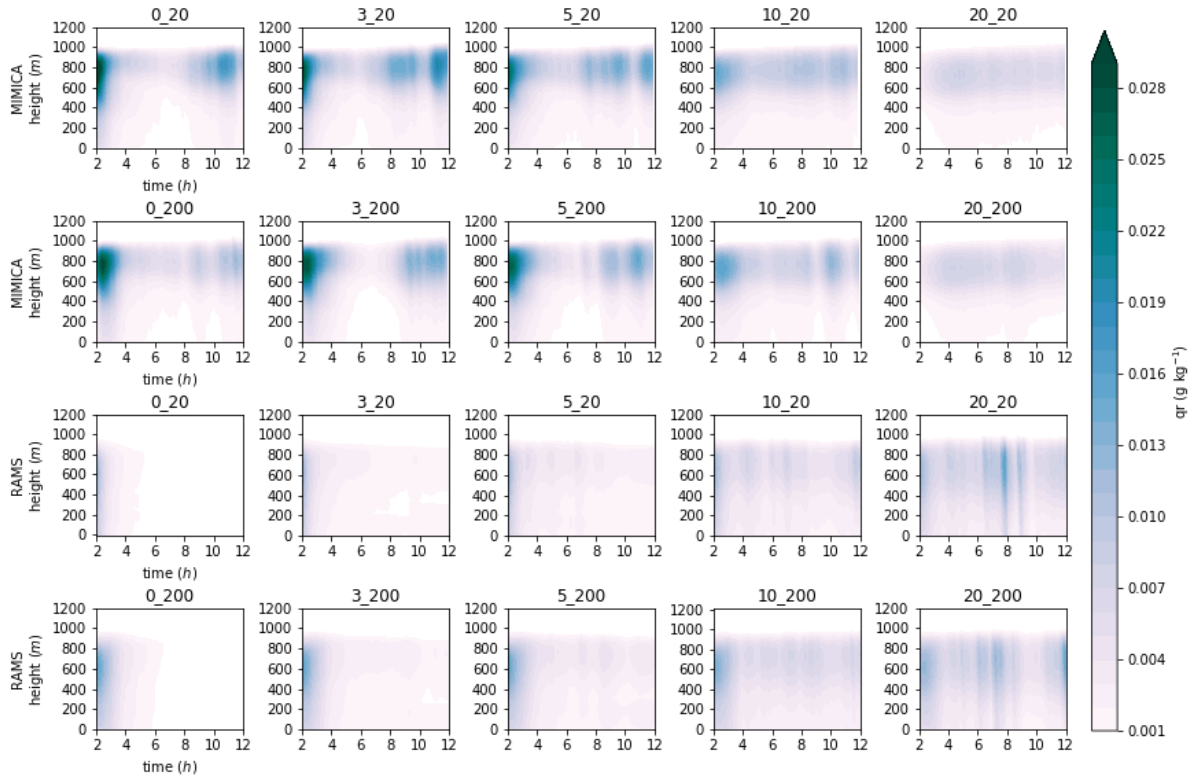
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plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "0_20" refers to "AC0_AK20". Black dashed lines represent the cloud top and cloud base heights.

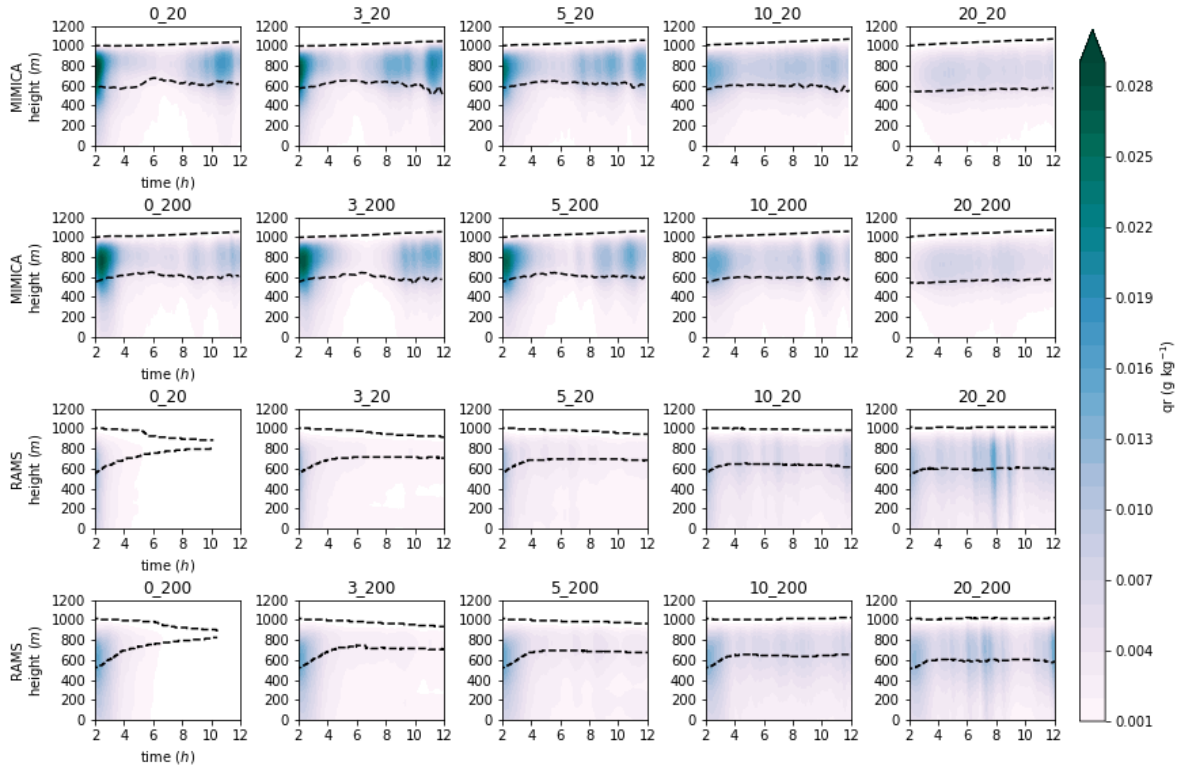


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Figure A3: Cloud top heights in (a) MIMICA and (b) RAMS. The first 2 h of simulations, considered as spin-up, are excludedThe first 2 h of simulations are excluded from the plots as they are considered as a spin-up period.



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Figure A4: Rain mixing ratio (qr) for the MIMICA and RAMS simulation sets. The first 2 h of simulations, considered as spin-up, are excluded ~~The first 2 h of simulations are excluded from the plots as they are considered as a spin-up period.~~ For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. “0_20” refers to “AC0_AK20”. Black dashed lines represent the cloud top and cloud base heights.

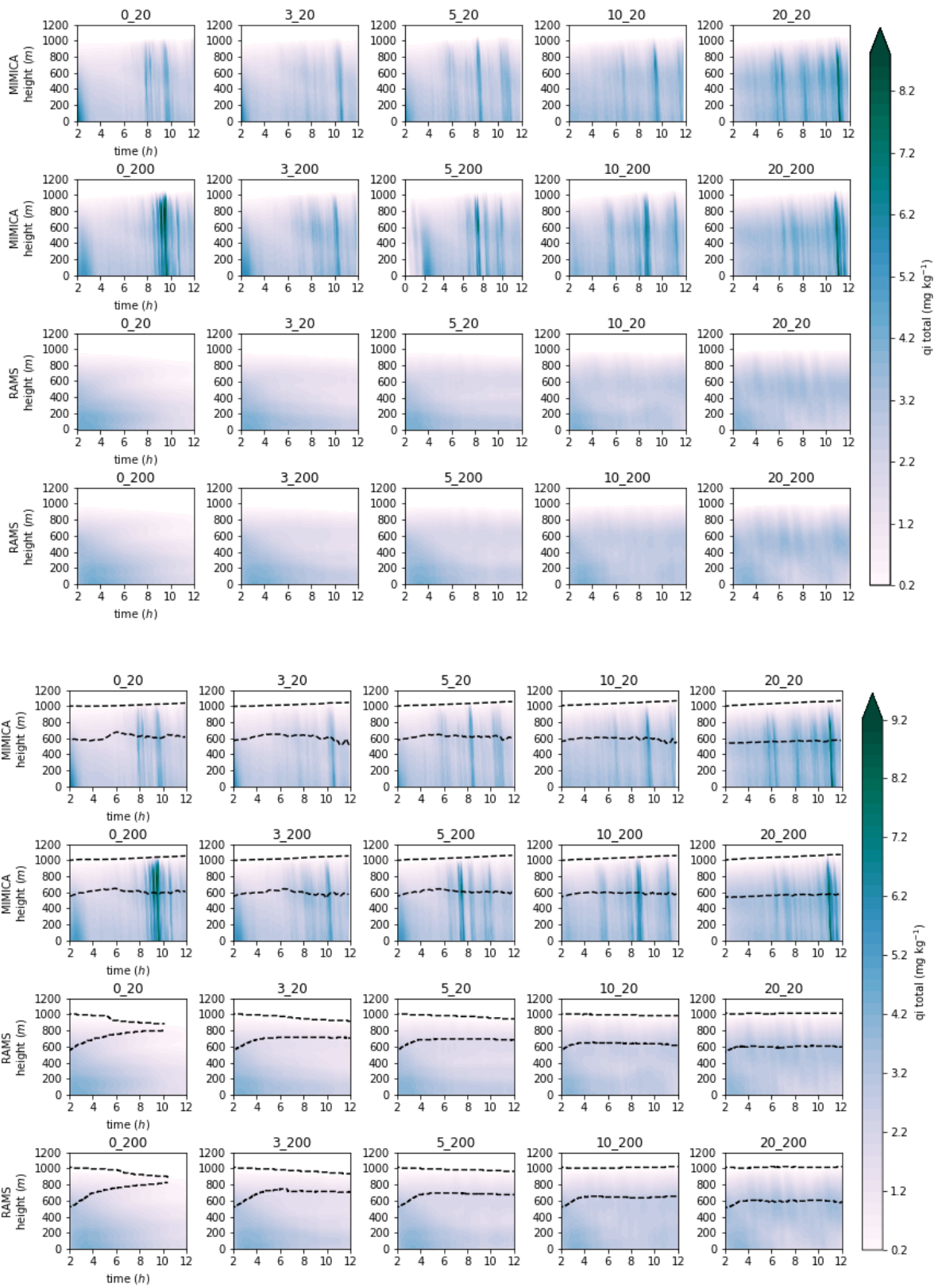


Figure A5: Total ice mixing ratio (q_i total) for the MIMICA and RAMS simulation sets. ~~The first 2 h of simulations, considered as spin-up, are excluded~~ The first 2 h of simulations are excluded from the plots as they are considered as a spin-up period. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation

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mode and the second to the Aitken mode concentration in cm^{-3} , i.e. “0_20” refers to “AC0_AK20”. Black dashed lines represent the cloud top and cloud base heights.

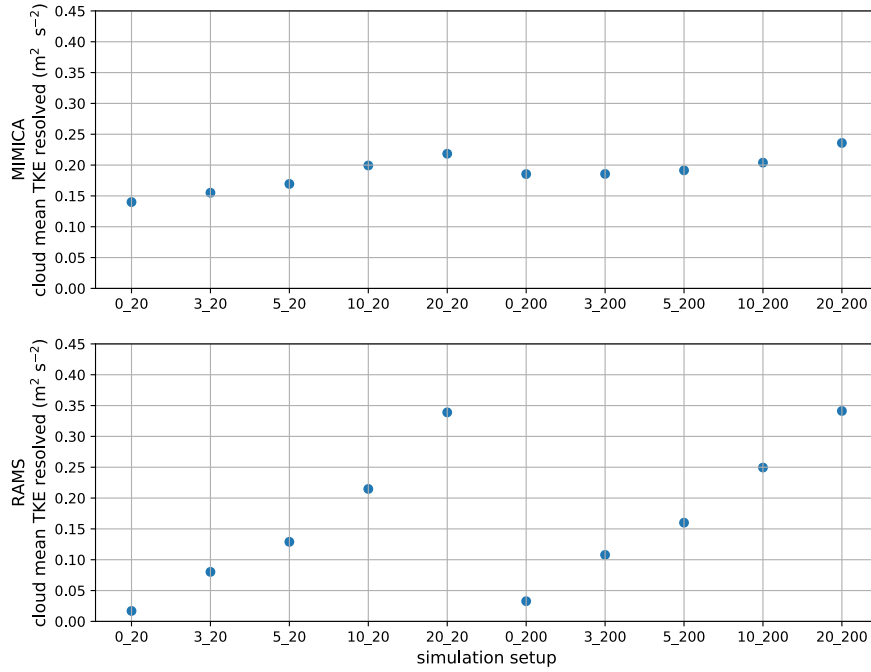


Figure A6: Time-mean resolved turbulent kinetic energy (TKE) averaged for the cloud layer, simulated by MIMICA and RAMS. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. “0_20” refers to “AC0_AK20”.

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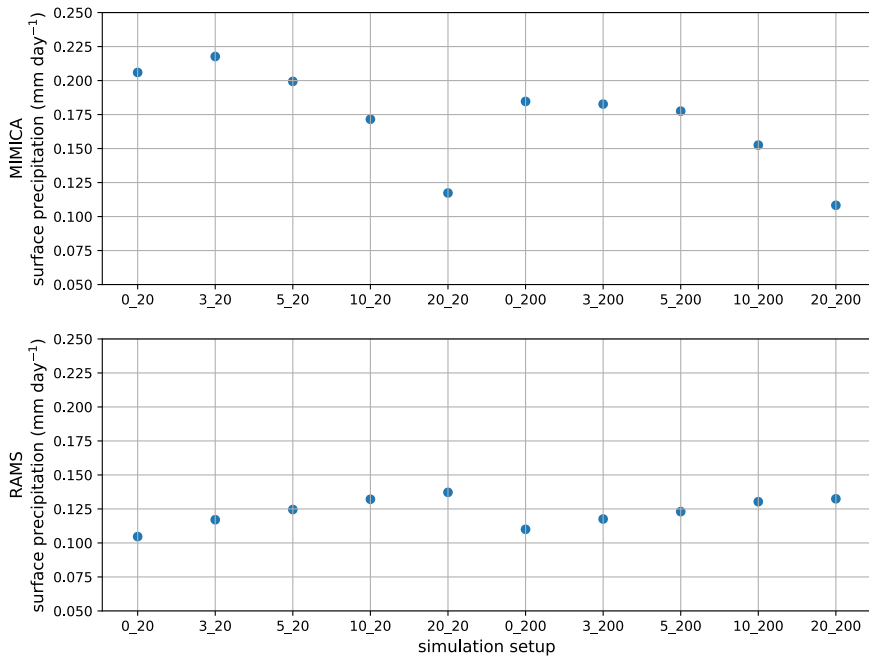


Figure A7: Time-mean surface precipitation simulated by MIMICA and RAMS. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm⁻³, i.e. “0 20” refers to “AC0 AK20”.

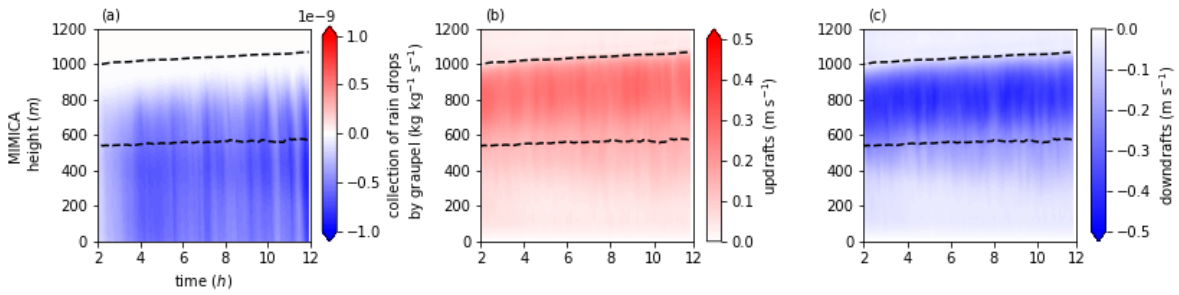
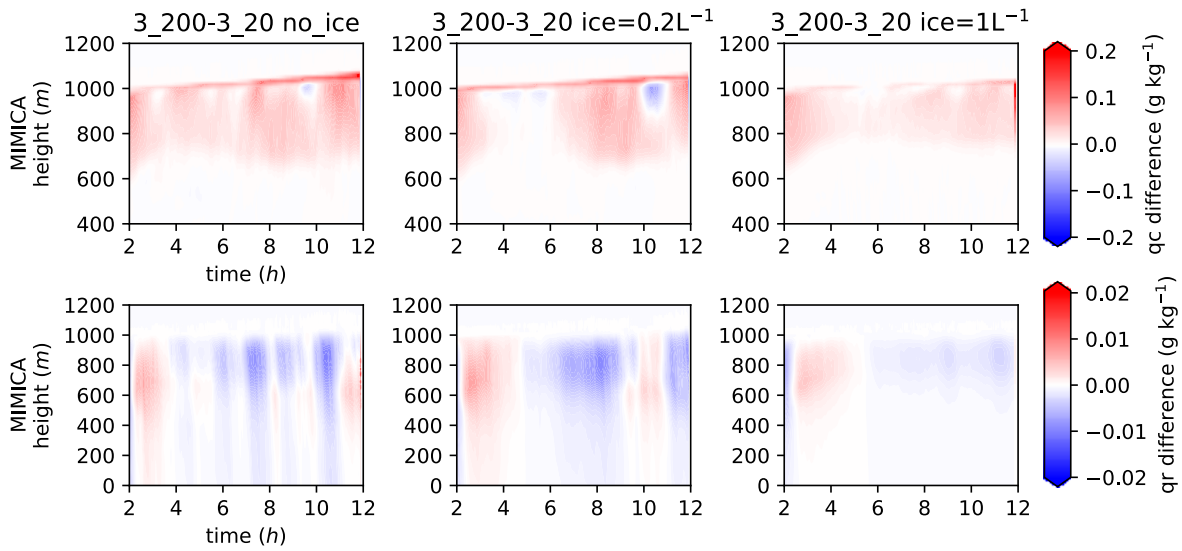


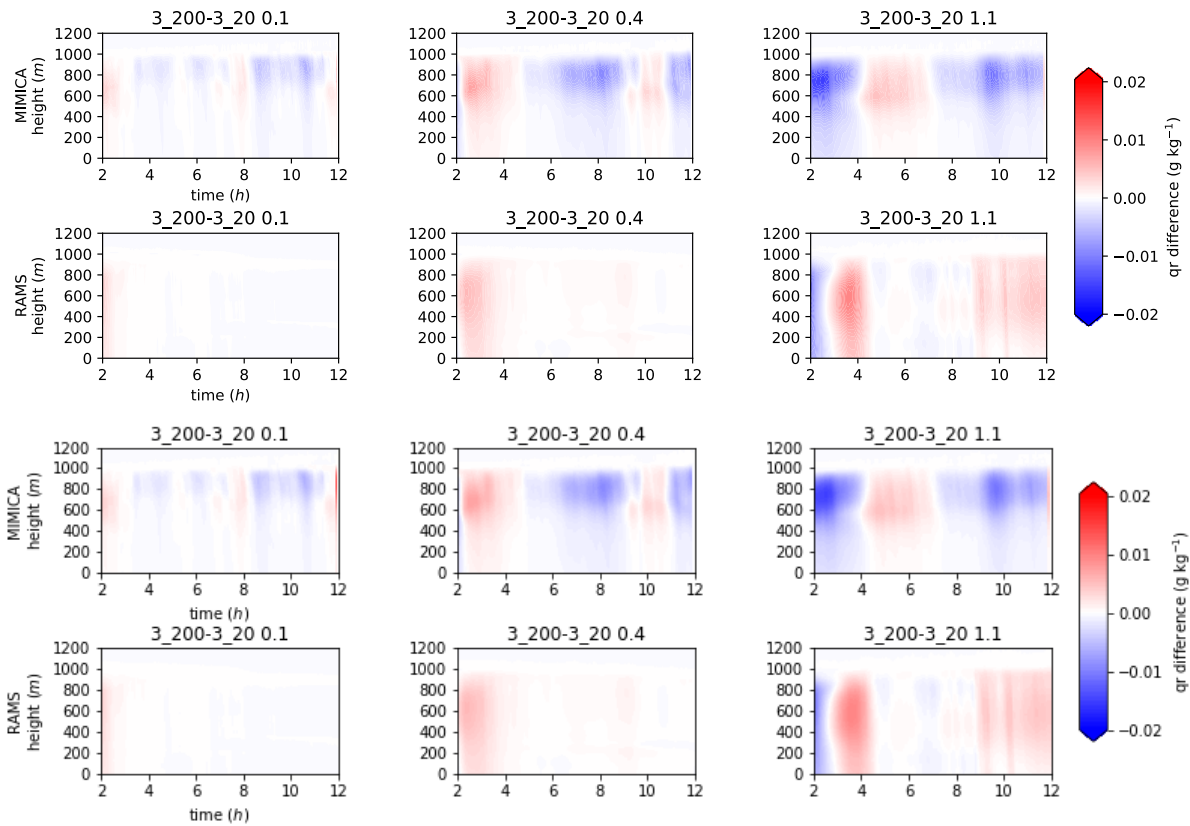
Figure A8: (a) Collection of rain drops by ice (b) updrafts (c) downdrafts with time, simulated by MIMICA. The first 2 h of simulations, considered as spin-up, are excluded. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm⁻³, i.e. “3 20” refers to “AC3 AK20. Black dashed lines represent the cloud top and cloud base heights.



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Figure A6: Differences in cloud-water mixing ratio (q_c) and rain mixing ratio (q_r) for simulation pairs with the same accumulation-mode concentration and the same ice crystal concentration: $=0 \text{ L}^{-1}$ (no_ice, the leftmost column); $=0.2 \text{ L}^{-1}$ (the middle column); $=1 \text{ L}^{-1}$ (the rightmost column) shown for MIMICA. The first 2 h of simulations are excluded as they are considered as a spin-up period. A student's t -test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "3_20" refers to "AC3_AK20".

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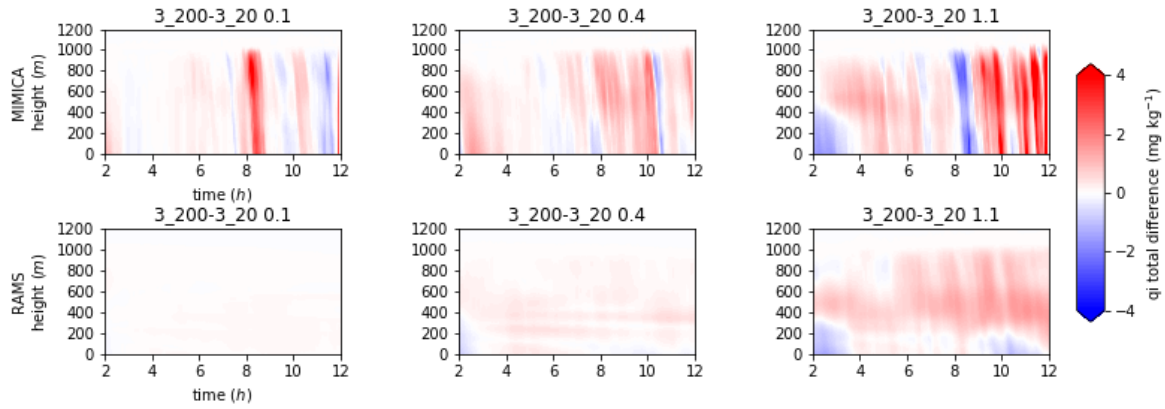
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Figure A97: Differences in rain mixing ratio (q_r) for simulation pairs with the same accumulation mode concentration and the same kappa value of the Aitken mode particles: $=0.1$ (the leftmost column); $=0.4$ (the middle column); $=1.1$ (the

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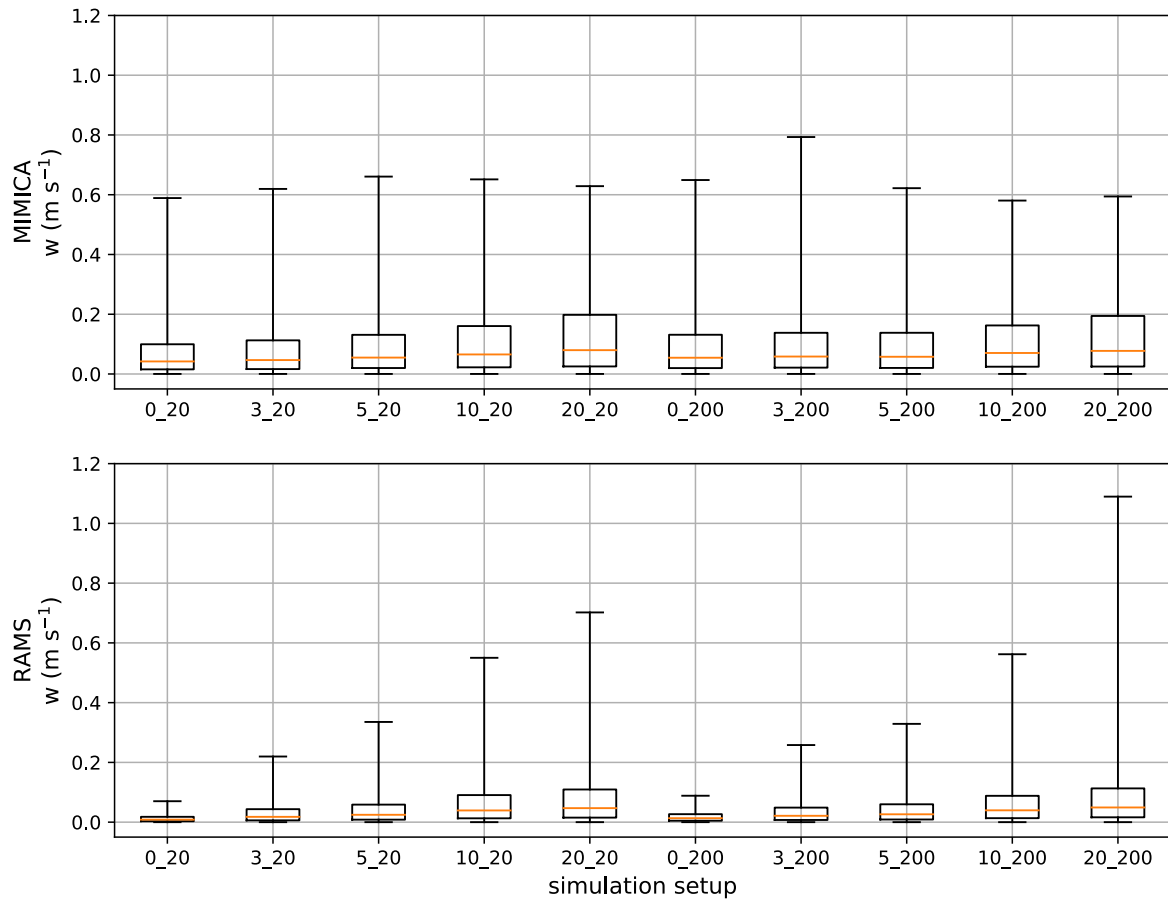
rightmost column) shown for MIMICA and RAMS. The first 2 h of simulations, considered as spin-up, are excluded~~The first 2 h of simulations are excluded as they are considered as a spin-up period.~~ A student's t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "3_20" refers to "AC3_AK20".



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Figure A108: Differences in total ice mixing ratio (q_i total) for simulation pairs with the same accumulation mode concentration and the same kappa value of the Aitken mode particles: =0.1 (the leftmost column); =0.4 (the middle column); =1.1 (the rightmost column) shown for MIMICA and RAMS. The first 2 h of simulations, considered as spin-up, are excluded~~The first 2 h of simulations are excluded as they are considered as a spin-up period.~~ A student's t-test with a 95% confidence level shows that the (time mean) differences are statistically significantly different for each pair of simulations. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. "3_20" refers to "AC3_AK20".

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1900 | **Figure A119:** Updraft (w) statistics ~~shown for a set of cases~~, simulated by MIMICA and RAMS. Lower and upper whiskers correspond to 1st and 99th percentiles, respectively. For figure clarity, the plot titles have been abbreviated; the first number refers to the accumulation mode and the second to the Aitken mode concentration in cm^{-3} , i.e. “0_20” refers to “AC0_AK20”.

1905 | Figure A120 shows the relationship between critical supersaturation and dry diameters calculated for a range of kappa values, i.e. $\kappa=[0.1; 0.2; 0.3; 0.4; 0.5; 0.6; 0.7; 0.8; 0.9; 1.0; 1.1]$. The computation is done for the temperature $T=298.15$ K and the surface tension $\sigma s/a=0.072$ Jm^{-2} . More details on the calculations can be found in Petters and Kreidenweis (2007).

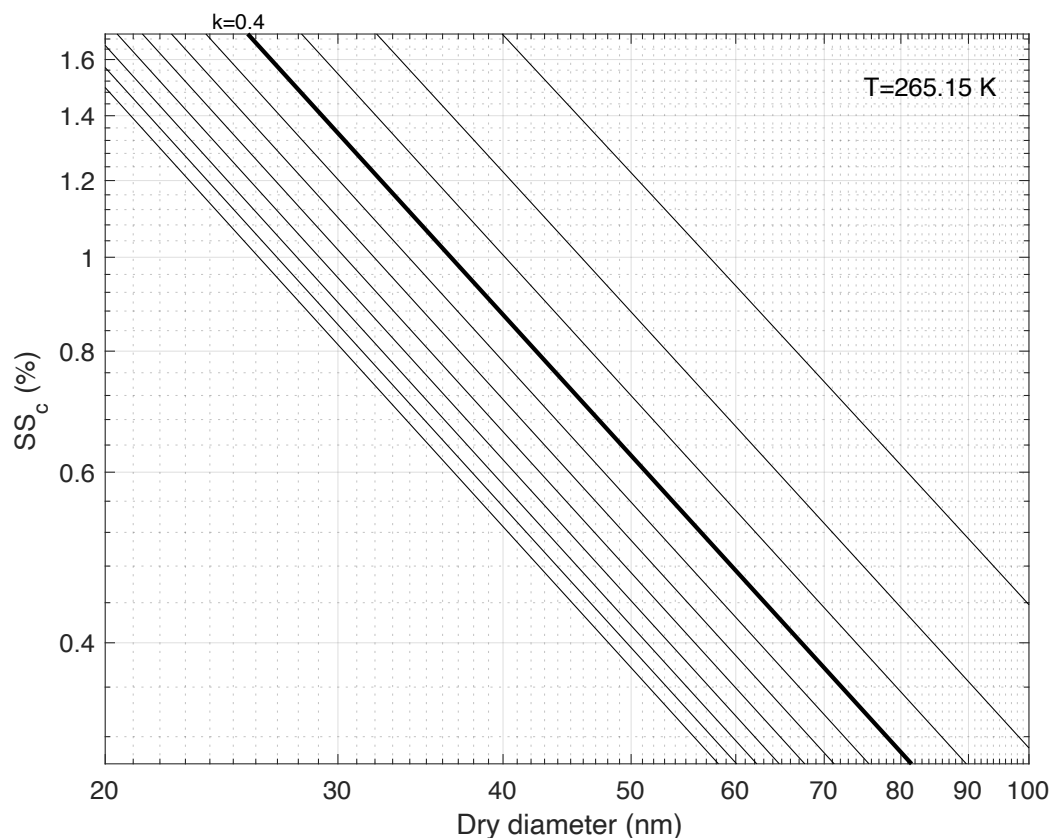


Figure A120: Calculated critical supersaturations SS_c (%) as a function of dry diameter, computed for $\sigma_s/a=0.072 \text{ J m}^{-2}$ and $T=298.15 \text{ K}$. κ -lines are shown for a range $0.1 \leq \kappa \leq 1.1$. Bold line corresponds to $\kappa=0.4$.

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Data availability. Modelling datasets used in this study are available at <https://bolin.su.se/data/bulatovic-2020> with assigned doi (<https://doi.org/10.17043/bulatovic-2020>). The observational datasets are available from corresponding authors upon request.

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Author contribution. IB, ALI and AMLE designed the experiments. IB and ALI performed the model simulations. IB analyzed the datasets. CL and JH provided the figures in the section “Qualitative comparison of model results with observational data for the High Arctic”. IB prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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