Response to Referee 1

We thank the referee very much for the many relevant comments. They helped to improve the paper considerably.

Note that we conducted new simulations because the ship emissions were shifted by two weeks in the old simulations. This especially affected the impact of transit ship emissions in the Arctic in late summer 2050. Furthermore, we increased the number of simulated years from 10 to 20 for better statistics, as suggested by referee 3. In some cases, the results have thus changed; as an example, the SW CRE now increases significantly with additional tenfold Arctic ship emissions in 2050.

Referee's comments in blue, our replies in *grey and italic*.

- In general, a lot of the discussion concerning specific results does not include the numbers in question and not references to where to find these numbers in the text (if present). This makes the discussion only qualitative. This is a shame, when the numbers are clearly available from the model results. Also, the reader ends up flipping back and forth looking for the numbers to back the discussion. Below, I will list some places in the text where this should be addressed.
Thank you very much for this comment. In the new version, we include now more quantitative information, namely numbers in brackets, additional Supplementary figures (Supplementary Figs. 4-14), and Tables 2 and 3.

- The reader lacks some of the basic information about the set up and the control simulation to be able to understand the results. There should, for instance, be a plot of the sea ice concentration at annual minimum or averaged over each season available for both periods, at least in the supplementary.
We include now plots averaged over each season for 2004 and 2050 in the paper (Fig. 2).

- A lot of the changes that occur between 2004 and 2050 are discussed, but not shown. This goes for example for temperature and precipitation. Make sure to label when the results you are referring to are not shown (see comments below) and please consider to show more of the changes that you use in your explanations, at least in the supplementary.
We included additional figures in the Supplementary Material (Supplementary Figs. 4-14) and now mention when something is not shown.

- For some figures, you average from 70-90N, for the tables you use either 60-90N or 75-90N. It would be more consistent if your figures and tables matched and one could follow the impacts of interest from figure to tables etc. Please consider changing this.
We now average the figures of concern (aerosol size distributions) between 75° and 90°N instead of 70° and 90°N to be consistent with the averages displayed in Tables 5 and 7, and with most averages given in brackets.

- Parts of the text is very oral and parts are too elaborate. Below I make both comments on things that should be changed and comments about how the text itself can be improved.

o P2, L24: "Nowadays" is very oral. Please rewrite. Also include " (. . .) pristine compared to other regions (. . .)" The use of depleted here make it sound like the aerosols have been removed. Please rewrite.
Changed to:
"Compared to other regions, the present-day Arctic air is exceptionally pristine, and aerosol levels are very low."

o P2, L32-P3,L8: May be a bit hard to follow because the indirect effects are described before the general radiative effects of clouds. Consider a rewrite to change the order.

We shifted the effects of aerosol-cloud interactions on radiation to the paragraph where the CREs are described.

o P3, L18: “Re-emission of SW”? Please rewrite!
Changed to:
“Aerosol scattering of shortwave (SW) radiation tends to cool the atmosphere, whereas absorption of SW and longwave (LW) radiation tend to warm it (Boucher et al., 2013).”

o P3, L23-27: Suggest to use the same terms for SW and LW description. For LW, the emissivity includes the water path and the temperature is height dependent. I suggest to make the definitions a bit more tidy.
Changed to:
“Similar to aerosol particles, clouds impact the Earth's radiation budget by absorption and emission of LW radiation (warming) and scattering of SW radiation (cooling). To a smaller extent, LW radiation is also scattered and SW radiation absorbed (Chou et al., 1999; Slingo 1989). The absorption and emission of LW radiation is a function of the emissivity of the cloud (which depends on microphysical cloud properties and the water path), the (height-dependent) cloud temperature, and the surface temperature (Corti and Peter 2009; Chen et al., 2006; Alterskjaer et al., 2010; Shupe and Intrieri, 2003). The scattering of SW radiation is a function of the microphysical cloud properties, of the cloud water path, of the solar zenith angle, and of the surface albedo (Corti and Peter, 2009; Liou et al., 2002; Shupe and Intrieri, 2003).”

o P4, L11: a bit confusing to have figure references in a listing of the main goals of the paper. Suggest to move this.
We moved the figure reference after the goals and changed the text to:
“Figure 1 provides a simplified overview of how the increase in Arctic temperature can affect radiation. The most important interactions between atmospheric variables, aerosols, clouds, and surface properties are included. The figure shows that the increase in temperature directly affects sea ice, specific humidity, and aerosols. Changes in these variables can then directly or indirectly impact clouds and radiation.”

o Figure 1: A bit confusing. Why not use red for increase and blue for decrease?
We adapted the figure following your suggestions.

o P5, L12: Does lowering the CDNC threshold affect the global radiative balance?
We used a model version that was retuned, which is now explicitly mentioned in the text.
Changes to:
“Thus, we decided to use 10 cm^{-3} as a lower threshold for the CDNC everywhere and retuned this new model version.”

o P8,L10: Define the abbreviations COADS and AMVER.
We did this for COADS. For AMVER, the acronym is better known than what it stands for, and the full name of AMVER has changed several times. Therefore, we rather refer to the homepage with a footnote.

o P9, paragraph starting on line 18: I find this very confusing and suggest a clarification of what you mean in this paragraph. Do not see the link between these two sentences. The reasoning therefore fails.
o P9, L25: Please rewrite to “(. . .) can be considered as a realistic (. . .)”
o P9 General comment to the justification of a tenfold increase of the ship emissions. I suggest a rewrite of this discussion. You increase emissions so as to see a signal and try to justify it afterwards, while at the very end of the paragraph state that the emissions are now “probably too high”. Increasing your emissions to get a signal is fine. To discuss that emission estimates may be too low is fine. Your emissions may well be an upper estimate. However, the discussion is long, a bit vague and a bit on the defensive side and makes the reader question whether the authors question their own reasoning here.

We rewrote the text to make it clearer (page 8 in the new document). We tried to account for all of your suggestions.

o P9, L30: Please define your abbreviations and give references to the models used.
Did you use RCp8.5 for future simulations here? Please specify.
Changed to:
"Both SIC and SST are prescribed in ECHAM6-HAM2. For future conditions, we used model results from the Earth System Model MPI-ESM as input (simulation for the climate model intercomparison project phase 5 (CMIP5), RCP8.5; Giorgetta et al., 2013)."

o P10, L9: The results would have appeared more robust if the sic and SSTs used
were an average taken over eg a ten year time period centered at 2004 and 2050.
Using one year (2003) and one ensemble (2050) to test the robustness of your choice
of sic and sst is too week.
We agree that the interannual variability in SIC and SST is large and only looking at one other ensemble might not be sufficient to confirm that our results are robust. However, we refrained from averaging SST and SIC over ten years since we wanted to conduct simulations with a realistic state of the Arctic Ocean. By averaging over SIC, less regions are either ice-free or totally covered with ice; instead, more regions with intermediate sea ice coverage exist, which can have impacts on aerosol emissions and clouds. Furthermore, we are not sure how much sense it makes to average SST in regions which are sometimes ice-free and sometimes covered by sea ice.

Changed to:
"We refrained from averaging SIC and SST over several years (e.g. 2000-2010) to avoid having spurious regions with intermediate SIC and SST. However, the interannual variability in SIC is pronounced, therefore we performed test simulations using SIC and SST from: i) the years 2003 and 2004 from AMIP and ii) the first and the second ensemble members from the MPI-ESM CMIP5 simulation for the year 2050. Overall, the Arctic SIC in 2003 was somewhat smaller than in 2004, and the SIC in the first ensemble member from MPI-ESM was smaller than in the second ensemble member. We found that the basic results and main conclusions do not depend on these differences in SIC but looking at only two years for both present-day and future might not be sufficient to confirm that all our results are robust. In the following, we will always refer to the simulations using SIC and SST from 2004 and future SIC and SST from the first ensemble member of MPI-ESM."

o P11,L10: reference needed after statement "Furthermore, most models (. . .) prevalent in the Arctic".
Changed to:
"Furthermore, models of different types generally have problems to reproduce the structure of mixed-phase clouds prevalent in the Arctic (Morrison et al., 2009; Klein et al., 2009; Fan et al., 2011; Morrison et al., 2011; Possner et al., 2017), and the future sea ice extent as well as the prescribed aerosol emissions are highly uncertain (Collins et al., 2013)."

o P11,L17: When you say "significantly" here is there a statistically significant change?
Please specify.
Yes. Supplementary Fig. 4 shows the changes. Moreover, we added the following sentence at the end of 2.4:
"Throughout this paper, the term "significant" is interchangeable with "statistically significant"."

o P11,L18: How much does the SIC decrease? Please specify.
We added the figures showing SIC (Fig. 2).

o P12,L3: "(. . .) modified (. . .)" What does modified mean here? Consider removing.
It refers to the fact that the sea salt parameterisation by Long et al. 2011 has been extended with corrections for SST, as mentioned in Section 2.1.2.
Changed to:

"Present-day emissions are a factor of ≈ 3 lower in our simulations compared with Browse et al. (2014), which results from the differences in the two parameterisations (Gong, 2003; Long et al., 2011, with SST corrections) as shown in the study of Long et al. (2011).

o P13,L5: Is it only the change in CCN concentration that affects CDNC? Not moisture availability? At what supersaturation do you calculate your CCN concentration? Does the average supersaturation change between the runs?
Thank you very much for pointing this out. We checked and found indeed that not only the changes in aerosol particles, but also changes in meteorology are responsible for the increases in CDNC. In our model, the CDNC depends on the calculated CCN concentration and the lower threshold of 10 cm^{-3}. The CCN concentration is calculated interactively following Köhler theory (parameterisation of Abdul-Razzah and Ghan, 2000) based on the aerosol size distribution and the maximum supersaturation. The maximum supersaturation depends e.g. on the updrafts, the temperature, and the CCN number concentration. We found that the updrafts available for activation increase between 75° and 90°N below ~750 hPa in early autumn (Supplementary Fig. 7), which contributes to the enhancement in CCN concentration.
Changed to:
"In general, the number of aerosol particles acting as CCN increases in the future, which leads to enhanced CDNCs (Fig. 4d). The increase in the number of CCN is not only caused by the increases in oceanic aerosol emissions, but also by changes in meteorology: the updrafts available for activation increase in the boundary layer between 75° and 90° N in early autumn (Supplementary Fig. 7), which supports the formation of cloud droplets in this region."

o P13, L22: Please explain why the increase in ICNC near the surface is due to the increase in CDNC.
This explanation was wrong. We thought that the increase in CDNC (as well as the increase in droplet radius) increases the contact freezing rate, but this is only important in limited areas far north. The simulated increases in ICNC are due to enhanced convection.

Changed to:
"The increase of ICNC near the surface is mainly caused by enhanced convection, which leads to small but numerous simulated ice crystals following the temperature-dependent empirical parameterisation of Boudala et al. (2002)."

o P13,L25: insert (not shown) after (near Svalbard).
We show this now in Supplementary Fig. 9. Furthermore, we correct "where precipitation is most enhanced" to "where convective precipitation is most enhanced" (which dries out the atmosphere and thus decreases cloud cover).

o P16,L2: ((. . .) except over the Arctic Ocean (. . .)" This is over the sea ice?! This should most definitely be specified.
Correct. Changed to: "except over sea ice"

o P16,L6: "since the clouds (. . .), more SW radiation can be absorbed (. . .)". Consider rewriting this for clarity.
Changed to:
"Part of this warming might be caused by BC and dust aerosols above clouds (Supplementary Fig. 10): the clouds reflect more SW radiation than the snow/ice-free surface and part of the scattered SW radiation can also be absorbed by aerosol particles causing an increase in aerosol absorption as compared to clear-sky conditions (see e.g. Myrhe et al., 1998)."

o P16;L13: What does the optical thickness change from and to? If you do not give the numbers you need to specify that it is not shown.
We added the numbers in brackets and in Tables 2 and 3.

o P17,24: Please insert "(not shown)" after "temperature".

We added the figure for surface temperature changes in early autumn to the Supplementary Material. Furthermore, the surface temperature is included in Tables 2 and 3 and now further discussed in the text. The text is changed to:
"This is because i) the SW component dominates in these months due to the higher zonal zenith angle and ii) the surface temperature over the central Arctic Ocean does not show pronounced increases like in early autumn (Table 2), therefore not enhancing the LW CRE. The surface temperature even decreases in some regions because melt ponds on ice can have temperatures higher than 271.38 K (but below 273.16 K) in late summer, while the SST is 271.38 K in gridboxes with 0<SIC<1 (equilibrium conditions, i.e. heat changes lead to changes in SIC, not SST)."

o Table 2: Please include the values from the control run to get the relative importance.
We now include the values from the control runs. Moreover, we splitted Table 2 and Table 3 (in the original document) for better readability into two tables each, one describing differences due to natural changes (Tables 4 and 5 in the new document) and one describing changes due to ship emissions (Tables 6 and 7 in the new document).

o P22, section 3.2.1 general comment: Your hypothesis is very likely, but can you verify in your model that this is the case? Perhaps you could perform some sensitivity test? Right now this section is not very strong as it just lists model results without any proper discussion. It would also be good to include a vertical cross section of the aerosol change. This would be beneficial for the next section as well when discussing freezing.
We added Supplementary Fig. 13 to strengthen our argumentation and discuss it in the text. Changed to:
"The additional aerosol particles emitted by ships provide additional surfaces for the condensation of gaseous sulphuric acid. Thus, the vertically integrated condensation rate of sulphate increases where the ship emissions occur (not significant; Supplementary Fig. 13b). The vertically integrated nucleation rate of sulphate shows neither a clear decrease nor a clear increase along the shipping paths (Supplementary Fig. 13d); if the increase in condensation suppressed nucleation, we would expect a decrease in the nucleation rate. However, the vertical cross section of aerosol particles in the nucleation mode shows that the number concentration indeed decreases significantly near the surface (Supplementary Fig. 13f)."

o P22,L29: How much does the Liquid water path /mass increase?
We now include the LWC in Fig. 11 (subfigures e and f).

o P23,L2: Please include the numbers you are describing in the text.
Changed to:
"Using satellite data, Christensen et al. (2014) studied the effect of ship tracks on both mixed-phase and liquid clouds. In the late summer of 2050, the clouds that are impacted by ships in our simulations are mostly liquid. Therefore, we restrict our comparison to the influence of ships on liquid clouds. Consistent with the observations by Christensen et al. (2014), we also found decreases in the effective radius and increases in cloud optical thickness. The relative changes in effective radius are larger in their observations (-20% at cloud top height) than in our simulations (-2% to -4% at altitudes below 500 hPa; averaged between 75° and 90° N, whereas changes in cloud optical thickness compare well (+20% in both studies, averaged between 75° and 90° N). The LWP slightly decreases in their analysis (-1%; in-cloud); in contrast, it increases in our simulations (+17%; all-sky, averaged between 75° and 90° N). While our simulated precipitation shows no clear trend, the results by Christensen et al. (2014) suggest that ship emissions delay precipitation by enhancing cloud lifetime. The different results could be explained by the location of the ship tracks analysed by Christensen et al. (2014): the majority of their samples lie between 45° S and 45° N, and only very few datapoints are from the Arctic. Precipitation formation at high latitudes differs considerably from that at low latitudes since e.g. convection is usually much more important at low latitudes."

o P25,L32: The significant areas are not large and looking at the figures it looks like the numbers you are giving here are averaged over the whole region. If so, please make it clear that this number is not only including significant changes.

We wanted to highlight with the given numbers that the local significant changes are small in absolute amount. We removed the numbers since they confused more than they helped. In the next sentence, it is mentioned that the changes in radiative forcing of BC deposition are much smaller than the changes in CREs.

o P25,L32: The figure reference should be to figure 13(f)?
Yes, indeed!

o P27,L8: "While the CRE (. . .)" is it not the change in CRE? Same goes for line 7 and the figure text to figure 14.
Yes, thank you.

o P27,L9: The change in optical thickness is significant in very small regions. This should be mentioned. Also when the average numbers are given in the following sentence, make clear that these numbers are not significant.
The changes are now significant and more widespread in the new simulations.
Changed to:
"While the changes in CRE caused by changes in cloud cover and cloud top altitude are not significant (Fig. 15a-d), the increase in cloud optical thickness leads to significant decreases and increases in the SW and LW CRE, respectively (Fig. 15e, f). Averaged between 75° and 90° N, the increased optical thickness changes the SW CRE by -4.6 W m^{-2} and LW CRE by 0.52 W m^{-2} in late summer (significant)."

o P27, L14: Are these numbers significant?
Yes; we now mention that.

Technical corrections: - Polarstereographic maps: Please insert a few more latitude lines, perhaps at the boarders for averages that you use: 70N and 75N.
We inserted every 5° a latitude line.

- Figures using a blue to red color scale: The lightest colors are impossible to separate in printed figures. You need to improve this color scale. At the same time, consider to use a white color surrounding zero so that values at this separation (zero values) do not come out in color.
We added a white surrounding around zero and adapted the color scales for many figures.

The following minor changes were adapted following the referees suggestions:
o P2, L17: Remove "where some (. . .) are labelled"
o P2,L20: Replace "until" with "before"
o P2, L22: Remove "(cruise ships)"
o P2, L28: Remove "and can (. . .) (Vali, 1985)" It is of no relevance here and only distracts the reader.
o P2, L32: Make part of the previous paragraph.
o P3, L1: Perhaps mention why smaller droplets increase the cooling effect of clouds?
o P3, L31: Suggest new paragraph before "How Arctic clouds (. . .)"
o P4, L22: Suggest to remove: "HAM2 (. . .) modes." and move "To link (. . .) implemented (. . .)" to the end of the next paragraph, after "(. . .) sedimentation".
o P6, L13: Suggested rewrite: "(..), we used an inventory described in (. . .)"
o P6,L19: Remove "more equations can be found therein"
o P8,L15: Remove "in addition". It is a bit confusing
o P8, L19: Please replace the word "exploit"
o P8, L21: Consider removing the sentence "We processed (. . .)". Too detailed information in my opinion.
o P9, L10: Suggest to remove "(non- (. . .) control)".
o P11, L3: suggest to remove "(eg. SW radiation, temperature)". It is not necessary.
o P11,L3: Rewrite to "change considerably"

o P11,L9: Rewrite to “deviate considerably”
o P11, L17: Rewrite “in the vicinity”. Suggest “Over the arctic ocean”
o P13,L3: Please insert the “(. . .) increases in the future (. . .)”
o P13,L8: “averaging over cloudy and non-cloudy conditions”. This is a bit confusing. Are you writing about allsky conditions? Also, please replace increases with increased.
o P16,L10: cooling effect vs warming effect. Please specify the actual numbers here.
o P16,L14: Consider replacing disentangle by “distinguish between”
o P22,L7-8: Please insert “(not shown)” after “late summer” and after “significant for OC”.
o P22,L12: Please remove “using the hypsometric equation”. Redundant.
o P22,14: Please move your reference to figure 9b to directly after “800 hpa” to avoid confusion.
o P25,L21: How much does the optical thickness change. You should strengthen your discussion by describing the actual model results.
o P25,L27: “(. . .) under clear-sky conditions.” Insert “(not shown)” here.
o P27,L4: please consider changing the numbers here to (-2 to -20 Wm-2)
o P27,L5: please insert “(not shown)” after “correlated with ship emissions”.
o P27,L16: Please insert “very”: “(. . .),ship emissions lead to a significant, but very weak (. . .)”
o P27,L20: Suggest to add “in limited regions” or something similar after “(. . .) and lead to significant net cooling”
o P32,L16: Insert “a very small” in front of “local warming”.
- All figures: Move (a), (b) etc above figures.
- Figure 7: Suggest to use different color scales for positive and negative values in (a) and (d).

Response to Referee 2

We thank the referee very much for his/her suggestions and comments, especially those concerning the impact of Arctic shipping. They helped to improve the paper considerably.

Note that we conducted new simulations because the ship emissions were shifted by two weeks in the old simulations. This especially affected the impact of transit ship emissions in the Arctic in late summer 2050. Furthermore, we increased the number of simulated years from 10 to 20 for better statistics, as suggested by referee 3. In some cases, the results have thus changed; as an example, the SW CRE now increases significantly with additional tenfold Arctic ship emissions in 2050.

Referee's comments in blue, our replies in *grey and italic*.

Page 1: cite Melia et al. 2016? (Melia, N., K. Haines, and E. Hawkins (2016), Sea ice decline and 21st century trans-Arctic shipping routes, Geophys. Res. Lett., 43, 9720–9728, doi:10.1002/2016GL069315.)
We added this interesting reference, which fits nicely into the introduction.

Page 3, line 18: "re-emission" – suggest rephrasing.
Changed to:
"Aerosol scattering of shortwave (SW) radiation tends to cool the atmosphere, whereas absorption of SW and longwave (LW) radiation tend to warm it (Boucher et al., 2013)."

Page 4, line 8-15: I think the motivation and objective for this study needs a couple of additional lines, e.g., to summarize what the bulk of the literature described above show about the importance of combining all the processes and what is new/unique about the present study.
We agree with the referee that more literature concerning the impact of ship emissions should have been mentioned and that we did not mention explicitly enough what is new in this study. We thus rewrote and extended the introduction (p. 4, l.23 to the end of the p. 4 in the new document).

Page 4, line 11: I don't see that this goal is sufficiently addressed in the paper. The model is run with fixed SSTs and no quantification of temperature responses. As far as I can tell, Fig.1 shows arrows only from temperature changes to radiative changes. If you want to maintain this as a main objective, you need to come back to it later in the manuscript in a better way. However, I think that disentangling the aerosol-radiation-cloud interactions is a sufficient objective in itself.
We agree with the referee. Since both SST and SIC are fixed, talking about temperature feedbacks is misleading. We can only refer to the impact on temperature by looking at the different radiative forcings. Therefore, we delete "Our goal is to draw conclusions about how changes in radiative forcings and radiative effects may feed back on temperature;"

Fig. 1: I like the figure, but find the colors a bit confusing. E.g., use of blue from less sea ice to more aerosols. Perhaps use red for increases and blue for decreases? Or add colors.
We changed the figure following the referee's suggestions (in line with the comment of referee 1).

Page 5, line 8: "simplistic treatment": please specify.
Changed to:
"This ensures that the global CDNC is not unrealistically low due to missing aerosol species in the model such as nitrate or due to the simplistic model description of organics (no explicit treatment of secondary organic aerosols; neglection of marine organics)."

Page 5, line 12: changing CDNC - does that affect the global radiative balance?
We used a retuned model version. This is now explicitly mentioned in the text.
Changed to:

"Thus, we decided to use 10 cm^{-3} as a lower threshold for the CDNC everywhere and retuned this new model version."

Page 5, line 12: is this based on observational constraints?
Only partly. It is a compromise between accounting for the low aerosol concentrations in the Arctic and missing aerosol sources in the model. We now included observational data of Arctic CCN/CDNC.
Changed to:
"In the standard ECHAM6-HAM2 setup, a minimum CDNC of 40 cm^{-3} is implemented. This ensures that the global CDNC is not unrealistically low due to missing aerosol species in the model such as nitrate or due to the simplistic model description of organics (no explicit treatment of secondary organic aerosols; neglection of marine organics). Without a lower threshold for CDNC, the model might underestimate the CDNC also in the Arctic, where organic aerosol particles are emitted from the sea surface microlayer (Hawkins and Russell, 2010; Bigg et al., 2004; Leck and Bigg, 2005; Chang et al., 2011). However, since the Arctic is a remote environment with low aerosol concentrations, observations show that the value 40 cm^{-3} is often undershot in this region: between July 15th and September 23th, Bigg and Leck (2001) measured daily median CCN concentrations between 15 and 50 cm^{-3} at a supersaturation of 0.25%. In July 2014, Leaitch et al. (2016) found a median CDNC of 10 cm^{-3} for low-altitude clouds (cloud top below 200 m) and of 101 cm^{-3} at higher altitudes. In October 2004, McFarquhar et al. (2007) conducted aircraft measurements in single-layer stratus clouds and found averaged cloud droplet number concentrations of 43.6±30.5 cm^{-3}. Applying the standard CDNC threshold of 40 cm^{-3} would drastically reduce the influence of changes in the CCN concentration and therefore impede aerosol-cloud interactions. Thus, we decided to use 10 cm^{-3} as a lower threshold for the CDNC everywhere and retuned this model version. The studies by Bigg and Leck (2001) and Leaitch et al. (2016) indicate that values even below this lower threshold can occur. While these measurements are representative for a specific point, our model represents average values over a larger area (1.875° × 1.875°), which should be less variable than a point measurement. Nevertheless, we acknowledge that the threshold of 10 cm^{-3} could still be too high under certain conditions. In the model, this threshold is occasionally hit, e.g. over the central Arctic Ocean or in the subtropics. "

Section 2.1.2: to make the methods section easier to follow, I recommend combining all description of emissions into one paragraph. This will also reduce the need to refer to following paragraphs, which makes this section a bit hard to follow. Furthermore, are marine organic aerosol emissions included?
We followed the referee's suggestion and combined the sections. Marine organics are not included. We now include the following sentence:
"Marine organic aerosol emissions are not considered in this study."

Page 6, line 18: all BC particles? Only hydrophobic? And only ship, or also other anthropogenic particles? Please specify.
Changed to:
"In ECHAM6-HAM2, dust and BC particles (also those emitted by ships) can act as INPs in the immersion mode when transferred to the internally mixed mode."

Section 2.2: Arctic_2050 vs Arctic_2050_shipping: the difference is a bit unclear. Does the former have Peters et al. 2050 ship emissions, but without the x10? Shipping emission factors are described as being lower due to regulations, which is why I wonder. If so, comparing these two does not give the total effect of changes in ship emissions, but the effect the x10 increase? Please clarify.
The section about the ship emissions is indeed not clearly written, which leads to this misunderstanding. We reformulated the paragraphs about the ship emissions as well as Section 2.2. The difference between ***arctic_2050*** *and* ***arctic_2050_shipping*** *is that the first does not include the Arctic ship emissions by Peters et al. (2011), while the latter includes these emissions enhanced by a factor of 10.*

Table 1: would be useful to add references for the emissions as well.

This is a good suggestion, which certainly helps the reader. We extended Table 1 accordingly.

Page 8, lines 20-22: I think these two sentences are excessively detailed.
Sentences are deleted.

Page 9, line 26: first you justify the increase, then you say it is probably too high? Consider revising for clarity.
We rewrote this section (p.8, l.10-31 in the new document).

Page 9, line 27: is it possible to add a reference?
When looking for a reference, we found that this sentence is too speculative and therefore we deleted this sentence.

Page 10, line 14: "naïve stipling approach" is not good language. Does this refer to a standard student's t-test? Please clarify/change.
The naive stippling approach refers to the following: "with this approach, a significance test is calculated for every gridpoint and all gridboxes are stippled where the p-value is smaller than 5% (for a significance level of α=0.05)." The wording "naive stippling" is used in the study by Wilks (2016) and we reuse it in lack of a better expression.

Page 11, line 1: figures show, not "will show". Consider changing the language.
We changed this.

Page 11, line 17: consider providing numbers or showing results in a supplementary material.
We show now the changes in the Supplementary Material (Supplementary Fig. 4) and in Tables 2 and 3.

Page 11, line 18: perhaps I misunderstand the language, but isn't the increase in wind speed following the reduced SIC the main reason for the increased DMS and sea salt emissions, and hence for the burden increase? Or are there other mechanisms, related to e.g., scavenging due to lower SIC that dominate the burden change? Please clarify.
The simulated sea salt emissions are a function of SIC, which acts as a barrier between the ocean and the atmosphere. At SIC=1, no sea salt and DMS is emitted from the ocean. In regions where SIC does not change, both wind speed and SST affect the emissions. We now again explicitly mentioned this in the result section.
Changed to:
"Over the central Arctic Ocean, the decrease in SIC (Fig. 2) enables emission fluxes of DMS and sea salt, which significantly increase their burdens (Supplementary Fig. 4; Tables 2, 3). As a second-order effect, significant increases in u_{10} (Supplementary Fig. 5) over the central Arctic Ocean in early autumn increase sea salt and DMS emissions. In regions where the SIC does not change, both (insignificant) changes in u_{10} and changes in SST (Supplementary Fig. 6) affect DMS and sea salt emissions, and thus their burdens. For example, the decrease in the sea salt burden over the Bering Strait is due to the decreases in SST (caused by a model bias in the MPI-ESM sea surface temperature compared to AMIP) and u_{10}."

Page 11, line 26: caused by what?
Since the precipitation increases, also the wet deposition is enhanced, which is the most important removal process for BC and OC in the Arctic in our model. (All BC and OC emissions are the same for the two simulations.) We now mention this in the paper.
Added the following sentence:
"The smaller BC and OC burdens can be explained by the increase in precipitation, which leads to enhanced wet deposition (the BC and OC emissions are identical between the two simulations)."

Page 11, line 32: JJA/August – do you consider a different period here? Please clarify.
Yes, for a fair comparison, we look at the same periods as the other two studies. We rewrote the text to make it clearer (p.12, l.28).

"When we compare our results to other studies, we average over the same time and space as they do for a fair comparison."

Page 12, line 1: at some point it would be good to show/describe in detail the changes in variables such as SIC between 2004 and 2050. Could be added in a supplementary material.
We added the figures showing SIC to the main paper (Fig. 2).

Page 12, line 4: absolute emissions in 2004 or absolute emission changes? Please clarify.
Changed to:
"The absolute present-day emissions..."

Section 3.1.2: are the same general features seen during summer?
Yes. If the season is not specified, (qualitative) results refer to both late summer and early autumn, as mentioned in the paper. We now provide more quantitative information in the text for the two seasons. Furthermore, we included two new tables (Tables 2 and 3), which also provide more quantitative information for the two seasons.

Page 16, line 13: again, it would be helpful to have the actual numbers.
We added the numbers.

Page 17, line 4: perhaps instead say "a strengthening of the direct aerosol effect" since it is in fact much stronger in 2100?
Page 17, line 5-7: I'm not convinced it makes sense to compare these numbers since the foundation and model experiments are so different. Unless you're able to disentangle effects of experimental differences in more detail, I don't see that this section add much information of value and it could be left out.
We agree with the referee and take this comparison out of the paper (this corresponds also to the referee's comment above).

Figure 5: very hard to distinguish statistically significant areas.
We changed the stippling from points to lines for better visibility.

Page 17, line 8: please add numbers or relative change.
We added Supplementary Fig. 11.

Page 17, line 9: if I follow correctly, these results are still without any changes in anthropogenic aerosol emissions, so a small effect due to changes in BC deposition is to be expected, unless there are large changes in the scavenging. Could be useful to remind the readers of this. In fact, even under RCP8.5, anthropogenic aerosol emissions decline strongly through the century, which could perhaps reduce this forcing altogether.
This is correct. When discussing changes in the size distribution, we now remind the reader that the emissions are identical ("The smaller BC and OC burdens can be explained by the increase in precipitation, which leads to enhanced wet deposition (the BC and OC emissions are identical between the two simulations).") Moreover, we now mention that the anthropogenic aerosol emissions decline under RCP8.5.
Changed to:
"Also most prescribed aerosol emissions (excluding DMS terrestrial emissions, biogenic organic carbon emissions, and ship emissions) follow RCP8.5, which decline in most industrial sectors from 2004 to 2050."

Page 22, line 7: A comparison with previous work using the Peters et al. inventory (without the x10 enhancement) could be useful, e.g., Ødemark et al. 2012; Dalsøren et al. 2013.
Dalsøren, S. B., Samset, B. H., Myhre, G., Corbett, J. J., Minjares, R., Lack, D., and Fu-

glestvedt, J. S.: Environmental impacts of shipping in 2030 with a particular focus on the Arctic region, Atmos. Chem. Phys., 13, 1941-1955, https://doi.org/10.5194/acp-13-1941-2013, 2013. Ødemark, K., Dalsøren, S. B., Samset, B. H., Berntsen, T. K., Fuglestvedt, J. S., and Myhre, G.: Short-lived climate forcers from current shipping and petroleum activities in the Arctic, Atmos. Chem. Phys., 12, 1979-1993, https://doi.org/10.5194/acp-12-1979-2012, 2012.
Thank you very much for this suggestion. We now use the study from Dalsøren et al. (2013) for comparing our radiative forcings/effects in Sections 3.2.3 and 3.2.4.

Page 22, line 8: the maximum changes occur at the same location as the emissions; however, there are statistically significant increases over much larger areas. Should be specified.
Changed to:
"The maximum increases in burden (see Fig. 9b) occur at the same locations as the ship emissions, but significant increases can spread over a large part of the Arctic (see Fig. 9c), as shown for the example of BC."

Page 22, line 11-17: are these shifts large enough to have notable implications, e.g., for forcing? Possible to discuss to add some context?
The changes in the size distribution can have an effect on the radiative forcing. However, we find that the radiative forcing by aerosols hardly changes. In general, changes in the number size distribution can not only affect the aerosol radiative forcing, but also the number of CCN.

Page 25, line 18-19: actual magnitudes would be useful.
Numbers are now included.

Section 3.2.3: this section is missing a discussion of and connection to studies of the radiative forcing of shipping, both in the Arctic and overall to global impacts. This is important given that main conclusion of the study concern the negligible impact of shipping aerosol emissions. In particular, a discussion of the impact of shipping found in studies that do include explicit treatment of aerosol-cloud interactions and/or offline radiative transfer calculations could be important.
We now include a comparison with the work from Dalsøren et al., 2013 (p.33, l.10-25 and p.34, l.10-15).

Page 32, line 20-25: be careful about the phrasing of this conclusion, as it does not cover other effects of shipping emissions, such as NOx-induced ozone changes and CO2.
Thank you for this comment. We include now the sentence:
"Furthermore, this study does not account for ship-induced changes in greenhouse gases (e.g. O_3, CO_2), which are also important forcers (Dalsøren et al., 2013; AMAP Assessment, 2015)."

Response to Referee 3

We thank the referee very much for his/her comments and suggestions, especially those highlighting that the uncertainty from this study must be communicated more clearly. They helped to improve the quality of the paper.

Note that we conducted new simulations because the ship emissions were shifted by two weeks in the old simulations. This especially affected the impact of transit ship emissions in the Arctic in late summer 2050. Furthermore, we increased the number of simulated years from 10 to 20 for better statistics. In some cases, the results have thus changed; as an example, the SW CRE now increases significantly with additional tenfold Arctic ship emissions in 2050.

Referee's comments in blue, our replies in *grey and italic*.

Gilgen et al present a set of sensitivity studies with the atmospheric GCM ECHAM-HAM. The control simulation is driven by conditions approximately representative for present-day, three sets of differing boundary conditions are then computed: (a) increased sea surface temperatures and decreased sea ice cover, (b), in addition, changed aerosol emissions, and (c) in addition, further ship emissions. Each of the simulations is run for a short period of ten years. A large set of results is presented. The study is to a large extent based solely on the results of the one model and thus the hypotheses developed are strongly dependent on the chosen parameterisations. Very little comparison to data (for the control simulation) is presented. In one paragraph, the cloud radiative effects are compared to SHEBA data – from this it seems that the model has a very large bias. I believe it would be necessary for an improved paper to at least show some evidence that the model performs satisfactorily in the Arctic in comparisons to observations, before the results from the sensitivity studies can be considered meaningful.

We agree with the reviewer that the results of this study are uncertain, especially because they were conducted with only one climate model. Therefore, more information about the performance of the model in the Arctic will help the reader to put the results of this study into context. Thus, we added a comparison of some key variables in the Supplementary Information: AOT; BC and sulphate concentrations at the surface; cloud cover; LWP and IWP; cloud radiative effects (SW, LW, and net) at the surface and at the TOA. The AOT and the IWP are underestimated, whereas all other variables compare well (the bias in the SW surface CRE mentioned in the old paper version was likely caused by the local surface albedo of the observations; see Supplementary Information for more details). Despite the model bias in AOT and IWP, we consider our results meaningful: in our opinion, model biases do not imply that the simulated changes are necessarily wrong. Vice versa, also a model that compares perfectly to present-day observations can make wrong projections for the future. Furthermore, we want to stress that observations in the Arctic are sparse and sometimes also uncertain.

Next to the detailed comparison that we provide in the Supplementary Information (the most important results of which are mentioned in the paper), we added the following paragraph to the results section:

"Heterogeneous freezing is still an active field of research, and contradictory evidence exists concerning the ability of combustion aerosols to act as INPs (Kanji et al., 2017). Laboratory results suggest that soot starts initiating freezing at temperatures ≤-30°C (Kanji et al., 2017, Fig. 1-7). On the other hand, Thomson et al. (2018) found an increase in INP concentrations in ship tracks at higher temperatures. The increases were small at temperatures around −20°C, moderate at −25°C (≈ 0.5 L^{-1}; saturation ratio of 1.22), and quite pronounced at -30°C (≈ 2 L^{-1}; saturation ratio of 1.32). The ship plumes were measured near the port of Gothenburg (57.7° N, 11.8° E) in 2013 and 2014, and the meteorology in general represented climate conditions of the late-autumn maritime North. If ship exhaust (not necessarily the BC particles) can indeed induce freezing at higher temperatures than in the laboratory-based BC-parameterisation used in our model, the impact on

cloud ice could be larger than in our simulations, especially in early autumn when temperatures are colder."

At two instances, the results are compared to previously-published results for similar scenarios. It is astonishing how different the results are. A key hypothesis is that sea salt emissions may substantially increase with decreasing sea ice coverage. Fundamentally, this is no surprise, so the question is how large this could be quantitatively. Unfortunately the two other model studies reported are much more different from the model presented here than the change due to sea ice retreat (one model has a factor of 3 more, the other, a factor of 1000 less emission flux in present-day conditions). Also the radiative forcing due to aerosol-radiation interactions is very different between models – the model presented here has a substantially positive forcing, the other model, a negative one. Since such results are easily available from multi-model ensembles (CMIP5 or AEROCOM), it would be easy to put the model the authors use into context, much beyond the two studies cited.

Our main goal was to compare how future sea salt emissions might change due to sea ice retreat. Therefore, we initally only compared our values with studies that looked specifically at this question. To our knowledge, nearly no CMIP5 model calculates interactively emissions of sea salt, but we agree that other model intercomparison studies can give insight into the spread in sea salt emissions between different models. Therefore, we included results from the study of de Leeuw et al. (2011) and added the following sentences:

"Note that at the present state, sea salt emissions are highly uncertain and differ considerably between models: a comparison of 11 chemical transport and global climate models shows that the global annual mass emissions of sea salt lie in the range between 2.2 and 9.9 × 10^{12} kg yr^{-1} for 9 of the models; 2 models calculate higher emissions of 22 × 10^{12} kg yr^{-1} and 118 × 10^{12} kg yr^{-1} (de Leeuw et al., 2011). For comparison, our simulated value (1.3 × 10^{12} kg yr^{-1}) is on the same order of magnitude as most of these models, but lower than in all of them because the parameterisation does not account for the contributions from spume drops (Long et al., 2011). Our simulated absolute increases in sea salt mass emissions might therefore be underestimated."

In the conclusions, we highlight again the uncertainty:
"Compared with observations, our model has a low bias in AOT and cloud ice, which could impact the absolute changes in the radiative aerosol forcing and the CREs. Furthermore, when we compare our results with other modelling studies that looked at natural aerosol changes with declining sea ice, we find large intermodel differences, e.g. concerning sea salt emissions. This highlights that the results from this study – as from any climate model study projecting the future – are uncertain."

When it comes to the interpretation of the results, much is left for speculation. If the authors choose to have a pure modelling study, why don't they at least precisely clarify the processes that change? Why not budgets for changes of CCN, INP? A table that lists all relevant numbers (e.g. for the entire region, and split for open ocean and sea ice surfaces) as simulated for the different scenarios would be useful (emission fluxes, CCN, INP, cloud particle concentrations, LWP, IWP).

We agree that our result's section is somehow speculative. It is not straightforward to understand 'what is going on' in the model, and understanding every single result in detail would be beyond the scope of this study. The large advantage of using a climate model is that it accounts for many processes and their couplings. In this case, the main goal was to look at aerosol-cloud interactions and radiative forcings with a complex aerosol-climate model, which has the drawback that the results stem from a combination of processes that cannot easily be separated.
Unfortunately, we do not calculate intermediate variables such as CCN or INP concentrations. The model calculates CDNC and ICNC based on the size and chemical composition of the aerosol particles, temperature, and supersaturation.
However, we agree with the referee that a table showing averages over the whole Arctic region, open ocean, sea ice, and regions where sea ice has melted is very interesting. Thus, we included additionally Tables 2 and 3 in the paper, which show averages for sea salt and DMS burdens, AOT,

LWP, IWP, cloud cover, CDNC burden, and surface temperature. The results of these tables are discussed in the text.

Specific comments:
p3 l30 – this is not "generally" true, e.g. not in summer (as the following sentence correctly acknowledges)
Changed to:
"Therefore, the LW absorption of clouds becomes more important and can dominate the total cloud radiative effect depending on the specific time and location."

p3 l32 – but it is likely a small effect (Pithan and Mauritsen). What is the reference for the following sentence ("generally...")?
Changed to:
"How Arctic clouds and their radiative effects will change in the future is still an open question. Generally, both the SW and the LW cloud radiative effect (CRE) are expected to become stronger when more CCN are available (Mauritsen et al., 2011). However, compared to other temperature feedbacks, the contribution of changes in Arctic clouds might be small (Pithan and Mauritsen, 2014)."

P5 l12 – it would be good to report already here whether this threshold is hit, and, if so, how often. It would further be good to analyse whether indeed the lack of nitrate or organics is a major problem of this model for the Arctic.
This threshold is occasionally hit, e.g. in the subtropics and over the Central Arctic Ocean. We now also provide a comparison with observations, as suggested by referee 2.

Changed to:
"In the standard ECHAM6-HAM2 setup, a minimum CDNC of 40 cm^{-3} is implemented. This ensures that the global CDNC is not unrealistically low due to missing aerosol species in the model such as nitrate or due to the simplistic model description of organics (no explicit treatment of secondary organic aerosols; neglection of marine organics). Without a lower threshold for CDNC, the model might underestimate the CDNC also in the Arctic, where organic aerosol particles are emitted from the surface microlayer (Hawkins and Russell, 2010; Bigg et al., 2004; Leck and Bigg, 2005; Chang et al., 2011). However, since the Arctic is a remote environment with low aerosol concentrations, observations show that the value 40 cm^{-3} is often undershot in this region: between July 15th and September 23th, Bigg and Leck (2001) measured daily median CCN concentrations between 15 and 50 cm^{-3} at a supersaturation of 0.25%. In July 2014, Leaitch et al. (2016) found a median CDNC of 10 cm^{-3} for low-altitude clouds (cloud top below 200 m) and of 101 cm^{-3} at higher altitudes. In October 2004, McFarquhar et al. (2007) conducted aircraft measurements in single-layer stratus clouds and found averaged cloud droplet number concentrations of 43.6±30.5 cm^{-3}. Applying the standard CDNC threshold of 40 cm^{-3} would drastically reduce the influence of changes in the CCN concentration and therefore impede aerosol-cloud interactions. Thus, we decided to use 10 cm^{-3} as a lower threshold for the CDNC everywhere and retuned this model version. The studies by Bigg and Leck (2001) and Leaitch et al. (2016) indicate that values even below this lower threshold can occur. While these measurements are representative for a specific point, our model represents average values over a larger area (1.875° × 1.875°), which should be less variable than a point measurement. Nevertheless, we acknowledge that the threshold of 10 cm^{-3} could still be too high under certain conditions. In the model, this threshold is occasionally hit, e.g. over the central Arctic Ocean or in the subtropics. "

P6 l1 – it would be good to comment on the results of Eckhardt et al. (ACP 2015)
Thank you for this reference. We included it in the Supplementary Material when comparing simulated and observed SO_4.

p8 l6 – 10 years seem very little for small forcings
We increased the number of years to 20.

p12 top paragraph – what do these discrepancies by a factor of about 3000 imply for

the fidelity of the results in terms of sea salt emission changes?
Thank you for this comment. We added a comparison with de Leeuw et al. (2011) and mention now the uncertainty in the conclusions (see text above).

P13 l8 increased
Corrected.

p13 l22 – i.e. homogeneous freezing of droplets?
This explanation was wrong. We thought that the increase in CDNC (as well as the increase in radius) increases the contact freezing rate, but this is only important in limited areas far north. The simulated increases in ICNC are due to enhanced convection.

Changed to:
"The increase of ICNC near the surface is mainly caused by enhanced convection, which leads to small but numerous simulated ice crystals following the temperature-dependent empirical parameterisation of Boudala et al. (2002)."

P13 l23 – at con-
stant ICNC?
Changed to:
"Between 500 hPa and 200 hPa, the enhanced ice water content is linked to the increase in ice crystal radius, while the ICNC slightly decreases."

L13 l27 – indeed surface fluxes? Or rather simply moist adiabat changes?
The explanation in Abe et al., 2016 is the following: "Because of the reduced sea ice, a more extended open ocean area increased the latent and sensible heat fluxes from the ocean to the atmosphere. Along with the seasonal march, the decreased atmospheric temperatures increased the temperature gradient between the air and sea surface in October. Therefore, the fluxes from the ocean to the atmosphere were enhanced in October rather than in September."
In our study, most pronounced increases in cloud cover occur where sea ice has melted (Table 3) and where also the changes in latent and sensible heat fluxes are most pronounced (significant changes; not shown in the paper).

P16 l2 – it would be important to clarify whether this section refers to the radiative forcing by aerosol-radiation interactions only, or to the effective radiative forcing due to aerosol-radiation interactions, or whether it includes aerosol-cloud interactions.
We calculate the radiative forcing by calling the radiation scheme once with and once without aerosols, i.e. it is the radiative forcing by aerosol-radiation interactions only. We now remind the reader of this in the results section and highlight in the methods that our radiative forcing refers to all aerosols (not only anthropogenic aerosols). Furthermore, we now apply abbreviations from the newest IPCC report in our paper (e.g. "RF_{ari}" or "ERF_{aci}") for clarification and avoid the term "Twomey effect".
We added this sentence:
"As mentioned previously, the aerosol radiative forcing refers to the instantaneous effect of all aerosols on radiation (RF_{ari})."

P16 l4
– what are the absorbing components, and why is the positive forcing so large? P16 l6
The absorbing components are BC and dust. Since the warming is only prevalent in the presence of clouds, we assume that the scattering of aerosol particles is less important in the presence of clouds and that the warming of aerosol particles might be enhanced by the higher SW radiation reflection by clouds.
Changed to:
"If the presence of clouds is considered, aerosol particles warm the atmosphere also over Alaska and northeast Siberia (late summer) and over the whole northern Russia (early autumn; shown in Fig. 7e). Part of this warming might be caused by BC and dust aerosols above clouds (Supplementary Fig. 10): the clouds reflect more SW radiation than the snow/ice-free surface and

part of the scattered SW radiation can also be absorbed by aerosol particles causing an increase in aerosol absorption as compared to clear-sky conditions (see e.g. Myhre et al. 1998). Moreover, the scattering of aerosol particles could become less important in the presence of clouds, which increases the relative importance of aerosol absorption to extinction.

– it would be useful to demonstrate this at least in the supplementary material (since the authors write "not shown" it seems hey have the analysis at hand)
We added Supplementary Fig. 10.

p17 l5 – how is the coincidence of approximately the same reduction by 0.2 Wm-2 explained? Is the same thing happening in both models?
Based on the comment of referee 2, we take this comparison out. In contrast to Struthers et al. (2011), we look at all aerosol particles, which makes a comparison difficult.

P18 l13 – i.e. the effect is twice as large as observed? The authors should report this analysis as a table or similar.
We provide now a more detailed analysis in the Supplementary Material.

How important are future marine and shipping aerosol emissions in a warming Arctic summer and autumn?

Anina Gilgen[1], Wan Ting Katty Huang[1], Luisa Ickes[1,2], David Neubauer[1], and Ulrike Lohmann[1]

[1]ETH Zürich, Institute for Atmospheric and Climate Science, Switzerland
[2]Now at Stockholm University, Department of Meteorology, Sweden

Correspondence to: Anina Gilgen (anina.gilgen@env.ethz.ch)

Abstract. Future sea ice retreat in the Arctic in summer and autumn is expected to affect both natural and anthropogenic aerosol emissions: sea ice acts as a barrier between the ocean and the atmosphere, and reducing it increases dimethyl sulphide and sea salt emissions. ~~A~~ Additionally, a decrease in the area and thickness of sea ice could ~~in addition~~ lead to enhanced Arctic ship traffic, ~~e.g. to shorten the paths~~ for example due to shorter routes of cargo ships. Changes in the emissions of aerosol particles can then influence cloud properties, precipitation, surface albedo, and radiation. Next to changes in aerosol ~~particles~~emissions, clouds will also be affected by increases in Arctic temperatures and humidities. In this study, we ~~quantified~~ quantify how future aerosol radiative ~~forcing, aerosol-cloud interactions,~~ forcings and cloud radiative effects might change in the Arctic in late summer (July/August) and early autumn (September/October).

Simulations were conducted for the years 2004 and 2050 with the global aerosol-climate model ECHAM6-HAM2. ~~In~~ For 2050, simulations with and without additional ship emissions in the Arctic were carried out to quantify the impact of these emissions on the Arctic climate.

~~We found that aerosol number concentrations in the Arctic will generally increase in the futuredue to enhanced emissions of~~ In the future, sea salt as well as dimethyl sulphide emissions and burdens will increase in the Arctic. The increase in cloud condensation nuclei, which is due to changes in aerosol particles and meteorology, will enhance cloud droplet number concentrations over the Arctic Ocean ~~.~~ ($+10\%$ in late summer and $+29\%$ in early autumn; in-cloud values averaged between 75° and 90° N). Furthermore, both liquid and total water ~~content will increase~~ path will increase ($+10\%$ and $+8\%$ in late summer; $+34\%$ and $+26\%$ in early autumn) since the specific humidity will be enhanced due to higher temperatures and the exposure of the ocean's surface.

Changes in both aerosol radiative forcings and cloud radiative effects at the top of the atmosphere will not be dominated by the aerosol particles and clouds themselves but by the decrease in surface albedo (and by the increase in surface temperature for the longwave cloud radiative effect ~~). Due~~ in early autumn). Mainly due to the reduction in sea ice, the aerosol radiative forcing will become less positive ~~and the~~ (decreasing from $0.53\,\mathrm{W\,m^{-2}}$ to $0.36\,\mathrm{W\,m^{-2}}$ in late summer and from $0.15\,\mathrm{W\,m^{-2}}$ to $0.11\,\mathrm{W\,m^{-2}}$ in early autumn). The decrease in sea ice is also mainly responsible for changes in the net cloud radiative effect~~more negative, i.e.~~, which will become more negative in late summer (changing from $-36\,\mathrm{W\,m^{-2}}$ to $-46\,\mathrm{W\,m^{-2}}$). Therefore, the cooling component of both aerosols and clouds will gain importance in the future.

We found that future Arctic ship emissions related to transport and oil/gas extraction (Peters et al., 2011, ACP) will not have a large impact on clouds and radiation: changes in aerosol concentrations only become significant when we increase these ship emissions by a factor of ten. ~~The~~ However, even with tenfold ship emissions, the net aerosol radiative forcing shows ~~only small, non-significant~~ no significant changes. Enhanced black carbon deposition on snow leads to a locally significant but very small ~~warming~~ increase in radiative forcing over the central Arctic Ocean in early autumn (no significant increase for average between 75° and 90° N). Furthermore, the tenfold higher ship emissions increase the optical thickness ~~of low clouds and thus induce a small Twomey effect (cooling)~~ and lifetime of clouds in late summer ~~. This Twomey effect has~~ (net cloud radiative effect changing from $-48\,\mathrm{W\,m^{-2}}$ to $-52\,\mathrm{W\,m^{-2}}$). These aerosol-cloud effects have a considerably larger influence on ~~temperature~~ the radiative forcing than the direct ~~effect~~ effects of particles (both aerosol particles in the atmosphere and particles deposited on snow)~~, but it is more uncertain because of the large variability of clouds~~. In summary, future ship emissions of aerosols and their precursor gases might have a net cooling effect, which is small compared to other changes in future Arctic climate such as those caused by the decrease in surface albedo.

1 Introduction

~~Arctic temperatures increase~~ In the last decades, Arctic temperatures have increased approximately twice as fast as the global average temperature, ~~partly~~ e.g. due to temperature and ice-albedo feedbacks (Pithan and Mauritsen, 2014), changes in the Atlantic Ocean thermohaline circulation (Chylek et al., 2009), and the decline in European anthropogenic SO_2 emissions since 1980 (Navarro et al., 2016). This temperature increase has been leading to reductions in both Arctic sea ice extent and thickness for the last decades: for the period from November 1978 (start of satellite records) to December 2012, the Northern Hemisphere sea ice extent decreased by $3.8 \pm 0.3\%$ per decade (Vaughan et al., 2013). This decrease is more pronounced in summer and autumn than in winter and spring (Vaughan et al., 2013). Since global and thus Arctic temperatures will ~~most likely increase further~~ further increase in the near future, the Arctic is expected to become ice-free in late summer within the next several decades (Collins et al., 2013; McFarquhar et al., 2011).

Sea ice concentration (SIC) refers to the percentage of an area which is covered with sea ice. Ocean areas with high SIC have a larger surface albedo and reduced exchanges of heat, momentum, and gases between the ocean and the atmosphere than areas with low SIC (Vaughan et al., 2013). With an open Arctic Ocean, natural aerosol emissions will increase because more sea salt particles and more dimethyl sulphide (DMS; a precursor for sulphate aerosol particles) will be emitted (Browse et al., 2014). Under present-day conditions, emissions from the ocean are already an important aerosol source in some Arctic regions in summer: measuring aerosol particles with radii between ~~0.25~~ $0.25\,\mu\mathrm{m}$ to $10\,\mu\mathrm{m}$ in Svalbard ~~, Deshpande and Kambra (2014) identified sea spray particles as the main source for Arctic summer aerosol particles. Note that~~ (a map of the Arctic ~~, where some important land masses (such as Svalbard) and regional seas are labelled,~~ can be found in the Appendix~~(~~, see Fig. A1), Deshpande and Kambra (2014) identified sea spray particles as the main source for Arctic summer aerosol particles. In a modelling study, Struthers et al. (2011) found that sea ice retreat might increase the sea salt aerosol number emissions in summer by a factor of two to three ~~until~~ by 2100. ~~In addition~~

Presently, the contribution of Arctic shipping to aerosol radiative forcings within the Arctic is very small compared to other emissions (AMAP Assessment, 2015). However, sea ice retreat might ~~also~~ cause an increase in ~~anthropogenic~~ shipping aerosol emissions over the Arctic Ocean, since reduced summer sea ice enables ships to ~~pass through~~ cross the Arctic Ocean. Cargo ships could shorten their paths ~~, tourism (cruise ships) and fishery sectors~~ (Corbett et al., 2010; Melia et al., 2016), tourism could be expanded (Eckhardt et al., 2013), and the Arctic oil and gas production will likely be intensified ~~(Eckhardt et al., 2013; Peters et al. . Nowadays, the~~ (Peters et al., 2011). Compared to other regions, the present-day Arctic air is exceptionally pristine~~and depleted of aerosols~~, and aerosol levels are very low. Hence, increases in both natural and anthropogenic aerosol emissions might have a strong effect on cloud properties and radiation. Furthermore, deposition of black carbon (BC) on snow and ice lowers the surface albedo (Warren and Wiscombe, 1985) and therefore has the potential to accelerate sea ice retreat (Flanner, 2013).

Aerosol particles influence clouds e.g. by acting as cloud condensation nuclei (CCN) or ice nucleating particles (INPs). Freezing processes involving INPs are called heterogeneous freezing~~and can be subdivided into different heterogeneous freezing modes, among them contact freezing and immersion freezing (Vali, 1985); ~~; for a recent overview on heterogeneous freezing modes, see Kanji et al. (2017). The ability of an aerosol particle to act either as a CCN or an INP depends on its size and its chemical composition (Boucher et al., 2013).

Hence, both aerosol concentration and composition influence cloud properties substantially (Boucher et al., 2013): at a constant liquid water ~~amount~~content (LWC), an increase in the number concentration of CCN changes the cloud droplet number concentration (CDNC); it leads to more but smaller droplets, ~~thus increasing the cooling effect of clouds ("Twomey effect"; Twomey, 1974,~~ which increases the total surface area of the cloud. Since cloud droplets must reach a certain size before they form rain, this process may delay the formation of precipitation (Albrecht, 1989). On the other hand, an increase in aerosol concentrations could also lead to enhanced precipitation due to the presence of INPs, which reduce the required supercooling and/or supersaturation for ice initiation. An earlier freezing of some cloud droplets, followed by the Wegener-Bergeron-Findeisen process, may rapidly form cold precipitation (Lohmann, 2002). Aerosol-cloud interactions can affect cloud properties and the onset and/or intensity of precipitation further, as described e.g. in Lohmann and Feichter (2005); Jackson et al. (2012). In ~~the Arctic~~ Arctic mixed-phase clouds, observations suggest that ~~high aerosol concentrations decrease~~ the number of precipitating ~~particles~~ ice particles decreases by 1-2 orders of magnitude under polluted conditions when aerosol concentrations are high (Lance et al., 2011).

However, clouds are not only affected by aerosol particles. Increasing atmospheric ~~temperatures are~~ temperature is expected to shift the melting and the freezing levels – and thus also cloud ice – to higher altitudes. Additionally, higher temperatures will increase evaporation from the surface and, consequently, the available water vapour in the atmosphere. An open ocean further amplifies the increase in water vapour. Analysing satellite data from 2000 to 2010, Liu et al. (2012) found a negative correlation between sea ice extent and cloud cover over the Arctic Ocean, which was statistically significant and especially pronounced between July and November. Recently, Abe et al. (2016) showed with a coupled atmosphere-ocean model that enhanced heat and moisture fluxes resulting from the reduction in sea ice cover are indeed responsible for the simulated increases in cloud cover.

Both aerosol particles and clouds impact the Earth's radiation budget. Whether an aerosol particle predominantly absorbs or scatters radiation depends on its physical and chemical characteristics. Aerosol scattering of shortwave (SW) radiation tends to cool the atmosphere, whereas absorption ~~and re-emission~~ of SW and longwave (LW) radiation tend to warm it (Boucher et al., 2013). The sum of scattering and absorption is called extinction. Since the aerosol extinction (normalised by the aerosol mass) is generally largest when the size of the particle is comparable to the size of the wavelength, the SW effect is more important than the LW effect for the majority of atmospheric particles (Stier et al., 2007). However, for large particles such as dust or sea salt, LW effects can become relevant (Stier et al., 2007).

Similar to aerosol particles, clouds impact the Earth's radiation budget by absorption ~~and re-emission~~ of LW radiation (warming) and scattering ~~and absorption~~ of SW radiation (~~predominantly~~ cooling). To a smaller extent, LW radiation is also scattered and SW radiation absorbed (Chou et al., 1999; Slingo, 1989). The absorption and ~~re-emission~~ emission of LW radiation ~~depend on~~ is a function of the emissivity of the cloud (which depends on microphysical cloud properties and the water path~~,~~), the (height-dependent) cloud temperature, ~~cloud height, and the emissivity of the cloud (Chen et al., 2006; Alterskjær et al., 2010; Shupe and In~~ and the surface temperature (Corti and Peter, 2009; Chen et al., 2006; Shupe and Intrieri, 2003). The scattering of SW radiation is a function of the ~~number, size, and phase~~ microphysical cloud properties, of the cloud ~~particles~~water path, of the solar zenith angle, and of the surface albedo ~~(Liou, 2002; Shupe and Intrieri, 2003)~~(Corti and Peter, 2009; Liou, 2002; Shupe and Intrieri, 2003) . Since aerosol particles influence cloud microphysics, they also impact cloud radiative effects (CREs). With a higher CCN concentration at constant LWC, more radiation is scattered back to space and the cooling effect of clouds is enhanced. This is the so-called "Twomey effect" (Twomey, 1974, 1977), also referred to as radiative forcing due to aerosol-cloud interactions (RF_{aci}; Boucher et al., 2013). Furthermore, changes in cloud lifetime (e.g. delayed precipitation; "Albrecht effect"; Albrecht, 1989) also affect the CREs. Together with RF_{aci}, these adjustments are referred to as the effective radiative forcing due to aerosol-cloud interactions (ERF_{aci}; Boucher et al., 2013).

Compared with the global mean, the SW radiative effect of Arctic clouds is less important because of the large solar zenith angle and the high surface albedo (Alterskjær et al., 2010). Therefore, the LW absorption of clouds becomes more important and ~~generally exceeds the SW effect. Hence,~~ can dominate the total CRE depending on the specific time and location. Arctic clouds warm the planet in the annual average and show a net cooling effect only in summer (Walsh and Chapman, 1998).

How Arctic clouds and their radiative effects will change in the future is still an open question. Generally, both the SW and the LW ~~cloud radiative effect (CRE)~~ CRE are expected to become stronger when more CCN are available (Mauritsen et al., 2011). However, compared to other temperature feedbacks, the contribution of changes in Arctic clouds might be small (Pithan and Mauritsen, 201 . Palm et al. (2010) suggested that the overall effect of enhanced aerosol concentrations is to increase the net warming effect of Arctic clouds because LW radiation dominates in the long polar winter. In contrast, a modelling study of Alterskjær et al. (2010) found that the increase in anthropogenic aerosol emissions since pre-industrial times has led to larger changes in the annual Arctic SW (-0.85 $\mathrm{W\,m^{-2}}$) than in the LW (0.55 $\mathrm{W\,m^{-2}}$) CRE at the surface. However, their simulated LW radiation effect was approximately one order of magnitude smaller than suggested by the observation-based study of Garrett and Zhao (2006). Whereas Garrett and Zhao (2006) considered measurements from a specific location (near Barrow, Alaska) and analysed strong pollution events, Alterskjær et al. (2010) simulated the effect over the whole Arctic (defined as north of 71° N in

their study) under all conditions. Other explanations for the different results include model uncertainties, especially regarding cloud cover and thin cloud frequency (Alterskjær et al., 2010). For the Arctic summer, Mauritsen et al. (2011) showed that an increase in the number of aerosol particles can either decrease or increase the net CRE depending on the background aerosol concentration.

~~Using the global aerosol-climate model ECHAM6-HAM2~~Therefore, the future increase in both natural and anthropogenic aerosol emissions due to sea ice decline is expected to influence radiation both directly and indirectly. The following studies investigated the impact of future changes in either natural or anthropogenic aerosol emissions: Struthers et al. (2011, using the global aeroso and Browse et al. (2014, using the global aerosol microphysics model GLOMAP) analysed the influence of enhanced natural aerosol emissions on Arctic clouds in the future; we will discuss their findings in the comparison with our results. The impact of Arctic shipping on black carbon deposition on snow and ice by 2050 was studied by Browse et al. (2013), who found only a small contribution of BC from ships. Dalsøren et al. (2013) used the chemical climate model OsloCTM2 to study the impact of enhanced global and Arctic shipping in 2030. In their high growth scenario, O_3 had the largest impact on radiative forcing in autumn (August to October).

In this study, we aim to quantify ~~the~~ changes in future Arctic aerosol particles from both natural and anthropogenic sources enabled by sea ice reductions. Furthermore, we analyse changes in clouds and radiation, which are partly caused by these changes in aerosol emissions. ~~Our goal is to draw conclusions about how changes in radiative forcings and radiative effects may feed back on temperature; Fig.~~ We use the state-of-the-art global aerosol-climate model ECHAM6-HAM2, which allows us to study changes in Arctic aerosols and their impact on climate.

Figure 1 provides a simplified overview of ~~the~~ how the increase in Arctic temperature can affect radiation. The most important interactions ~~that can lead to possible temperature feedbacks.~~ between atmospheric variables, aerosols, clouds, and surface properties are included. The figure shows that the increase in temperature directly affects sea ice, specific humidity, and aerosols. Changes in these variables can then directly or indirectly impact clouds and radiation.

The model and the simulations, the boundary conditions, the emissions, and the used statistical method are described in Sect. 2. In the results and discussion section (Sect. 3), we ~~will~~ focus on the months July to October, when both the decrease in SIC and the increase in shipping through the Arctic Ocean will be most pronounced. In the conclusions (Sect. 4), our key findings ~~will be~~ are summarised.

2 Methodology

2.1 ECHAM6-HAM2

2.1.1 General information about ECHAM6-HAM2

ECHAM6-HAM2 is the combination of the general circulation model ECHAM6 (Stevens et al., 2013) with the two-moment cloud microphysics scheme by Lohmann et al. (2007) and the aerosol model HAM2 (Stier et al., 2005; Zhang et al., 2012). ECHAM6 solves prognostic equations for vorticity, divergence, surface pressure, and surface temperature and uses a flux

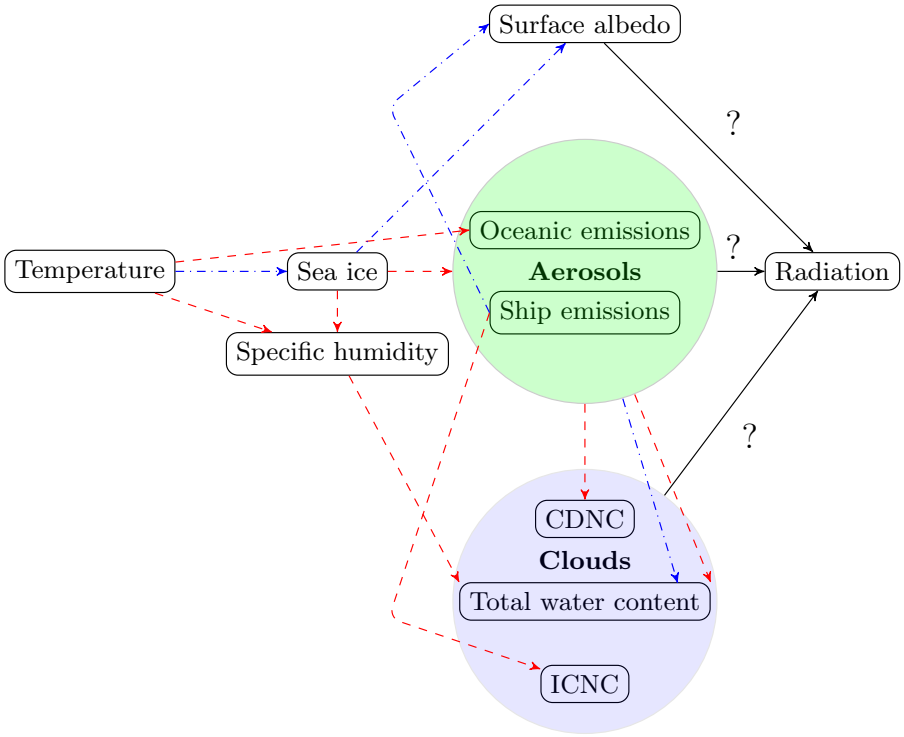

Figure 1. Simplified sketch showing how different variables (may) vary as a result of ~~changes in~~ enhanced Arctic temperatures. Red dashed arrows denote ~~enhancing effects ("increase → increase" or "decrease → decrease")~~expected increases, blue dashdotted arrows ~~dampening effects ("increase → decrease" or "decrease → increase"). For example, the future increase in Arctic temperature will lead to a decrease in sea ice concentration~~expected decreases. Black solid arrows show which components impact radiation. CDNC and ~~therefore again temperature~~ICNC stand for cloud droplet and ice crystal number concentration, respectively. Note that an increase in aerosol concentrations can either increase or decrease precipitation and thus the total water content, as mentioned in Sect. 1.

form semi-Lagrangian transport scheme to advect water vapour, cloud liquid water, cloud ice, and trace components. ~~HAM2 considers different aerosol species and different size modes. To link the simulated aerosol population with the CDNC and the ice crystal number concentration (ICNC), parameterisations for cloud droplet activation and ice nucleation were implemented (Abdul-Razzak and Ghan, 2000; Lohmann et al., 2008).~~

In HAM2, the aerosol components ~~sulphate~~SO_4 (sulphate), BC, organic carbon (OC), sea salt, and mineral dust are considered (Zhang et al., 2012). The size distribution of the aerosol particles is described by four size ranges: the nucleation mode ($r_m < 5\,nm$; r_m is the mode radius of the aerosol particles), the Aitken mode ($5\,nm < r_m < 50\,nm$), the accumulation mode ($50\,nm < r_m < 500\,nm$), and the coarse mode ($r_m > 500\,nm$). Only a soluble mode exists for the nucleation mode, whereas a soluble/internally mixed and an insoluble mode exist for the other three size modes. Therefore, seven aerosol modes are considered in total, each described by a log-normal size distribution. Coagulation and condensation can shift aerosol particles to larger modes and/or from insoluble to internally mixed modes. Removal processes of aerosol particles in ECHAM6-HAM2

comprise wet deposition, dry deposition, and sedimentation. To link the simulated aerosol population with the CDNC and the ice crystal number concentration (ICNC), parameterisations for cloud droplet activation and ice nucleation are implemented (Abdul-Razzak and Ghan, 2000; Lohmann and Diehl, 2006; Lohmann et al., 2008).

Regarding the sulphur chemistry, DMS is oxidised to SO_2 (sulphur dioxide), which can form sulphuric acid in the aqueous phase or in the gas phase. Gas-phase sulphuric acid in the atmosphere can either nucleate, i.e. form new small, soluble particles, or condense ~~on~~ onto pre-existing aerosol particles. Condensation can be limited by the available surface area of aerosol particles, by the available gas-phase sulphuric acid, or by the diffusion of the gas-phase sulphuric acid to the particle surface. If any gas-phase sulphuric acid is left after condensation, the sulphuric acid nucleates and forms new sulphate particles. Besides the available concentration of sulphuric acid, nucleation depends on temperature and relative humidity.

In the standard ECHAM6-HAM2 setup, a minimum CDNC of $40\,\mathrm{cm}^{-3}$ is implemented. This ensures that the global CDNC is not unrealistically low due to missing aerosol species in the model such as ~~ammonium~~ nitrate or due to the simplistic model description of organics ~~. Since~~ (no explicit treatment of secondary organic aerosols; neglection of marine organics). Without a lower threshold for CDNC, the model might underestimate the CDNC also in the Arctic, where organic aerosol particles are emitted from the sea surface microlayer (Hawkins and Russell, 2010; Bigg et al., 2004; Leck and Bigg, 2005; Chang et al., 2011) . However, since the Arctic is a remote ~~, aerosol poor environment , the value~~ environment with low aerosol concentrations, observations show that the value of $40\,\mathrm{cm}^{-3}$ is often undershot in this region~~(Bigg and Leck, 2001; Leaitch et al., 2016)~~: between July 15^{th} and September 23^{th}, Bigg and Leck (2001) measured daily median CCN concentrations between $15\,\mathrm{cm}^{-3}$ and $50\,\mathrm{cm}^{-3}$ at a supersaturation of 0.25%. In July 2014, Leaitch et al. (2016) found a median CDNC of $10\,\mathrm{cm}^{-3}$ for low-altitude clouds (cloud top below $200\,\mathrm{m}$) and of $101\,\mathrm{cm}^{-3}$ at higher altitudes. In October 2004, McFarquhar et al. (2007) conducted aircraft measurements in single-layer stratus clouds and found averaged cloud droplet number concentrations of $43.6 \pm 30.5\,\mathrm{cm}^{-3}$. Applying the standard CDNC threshold of $40\,\mathrm{cm}^{-3}$ would drastically reduce the influence of changes in the CCN concentration and therefore impede aerosol-cloud interactions. Thus, we decided to use $10\,\mathrm{cm}^{-3}$ as a lower threshold for the CDNC everywhere and retuned this new model version. The studies by Bigg and Leck (2001) and Leaitch et al. (2016) indicate that values even below this lower threshold can occur. While these measurements are representative for a specific point, our model represents average values over a larger area ($1.875° \times 1.875°$), which should be less variable than a point measurement. Nevertheless, we acknowledge that the threshold of $10\,\mathrm{cm}^{-3}$ could still be too high under certain conditions. In the model, this threshold is occasionally hit, e.g. over the central Arctic Ocean or in the subtropics.

2.1.2 Aerosol emissions

Emissions of sea salt, dust, and oceanic DMS are calculated online and depend on the $10\,\mathrm{m}$ horizontal wind speed (u_{10}). Marine organic aerosol emissions are not considered in this study. Sea salt emissions follow Long et al. (2011) with sea surface temperature (SST) corrections according to Sofiev et al. (2011). The correction is applied because SST affects sea salt emissions by influencing bubble rising velocities, the gas exchange between the bubbles and the water, the bubble bursting behaviour, and maybe also the coverage of oceanic whitecaps (Lewis and Schwartz, 2004). Dust emissions are calculated as stated in Tegen et al. (2002), with some modifications based on Cheng et al. (2008). The monthly mean DMS seawater con-

centrations are prescribed according to Kettle and Andreae (2000), and the flux from the ocean to the atmosphere is calculated following Nightingale et al. (2000). Changes in oceanic DMS concentrations are not straightforward to project: taking primary production or SST as a proxy seems not justified since Arctic oceanic DMS concentrations also depend on taxonomic differences in phytoplanktonic assemblages (Becagli et al., 2016). Using a coupled ocean-atmosphere model (with ECHAM5-HAM as atmospheric component), the study by Kloster et al. (2007) explicitly simulates DMS but only reports changes between the time periods 2061-2090 and 1861-1890, which are not directly comparable to the time periods we are interested in. Thus, we decided to leave the oceanic DMS concentrations unchanged.

Besides dust, sea salt, and oceanic DMS, the emissions of all other aerosol components or sulphate precursors are prescribed, mainly from the ACCMIP emission inventory (Lamarque et al., 2010). For ship emissions, we used ~~a different inventory, which is described in Sect. ??~~the inventories by Dalsøren et al. (2009) and Peters et al. (2011), which are described in the next paragraphs. Ship emissions are put into the second lowest model layer (~~≈ 150 m~~≈ 150 m). While OC and BC particles from ships are exclusively emitted into the insoluble Aitken mode, the sulphate mass is equally distributed between the accumulation and the coarse ~~mode~~modes. It is assumed that 2.5 % of SO_2 from ships is emitted as primary ~~SO_4 (sulphate; Dentener et al., 2006)~~sulphate (Dentener et al., 2006).

Our ship emissions are based on the inventories by Dalsøren et al. (2009) and Peters et al. (2011), which include the species SO_2, BC, and OC. The shipping emissions for the year 2004 follow Dalsøren et al. (2009), who combined the observational data sets COADS (Comprehensive Ocean-Atmosphere Data Set) and AMVER[1] considering ships above 100 gross tons. For the global ship emissions in the year 2050, we use the Dalsøren et al. (2009) ship emission inventory and apply the same reduction in emission factors for 2050 as in the study by Peters et al. (2011) (80% for SO_2 and 20% for OC), which are based on the Amendments to MARPOL Annex VI adopted by the International Maritime Organization in 2007.

For additional ship emissions in the Arctic in 2050, we take the ship emissions by Peters et al. (2011). They used the 2004-inventory by Dalsøren et al. (2009) as a "background" for calculating future Arctic ship emissions in the year 2050 for transit shipping and for shipping that is related to oil and gas production. Changes in ship emissions from the sectors tourism, fishery, and local/national transport are not considered. For the year 2004, no transit shipping was assumed, and the oil and gas shipping was estimated based on oil tankers operating in the Arctic region. The expected increase in these two sectors is related to SIC: less sea ice will faciliate the passage through the Arctic ocean and expose new areas to oil and gas production. Peters et al. (2011) assumed that emission factors of SO_2 and OC will decrease due to regulations and improved technology but that everything else (other aerosol emission factors; shipping routes) will remain constant.

We increased Arctic ship emissions by a factor of ten to detect a significant signal in aerosol particles. This is in agreement with the results of Peters et al. (2014), who studied the effect of ship emissions on tropical warm clouds with ECHAM5-HAM. In the following, we show how realistic these tenfold emissions are in the context of other studies and recent findings.

Compared with other estimates of future Arctic transit shipping, the results from Peters et al. (2011) lie between those from Paxian et al. (2010) and Corbett et al. (2010): the fuel consumption by Paxian et al. (2010) is 1.4 to 2.4 times smaller than the

[1] http://www.amver.com/

values reported by Peters et al. (2011). Depending on the scenario, the estimated CO_2 emissions by Corbett et al. (2010) are 2 to 4.6 times higher in 2050 than the values reported by Peters et al. (2011).

Recently, McKuin and Campbell (2016) pointed out that both global and Arctic ship emission inventories might underestimate BC ship emissions because too low BC emission factors were used. While the ship emission inventory by Peters et al. (2011) used a BC emission factor of 0.35, McKuin and Campbell (2016) found – depending on the averaging method and the area – factors between 0.79 and 0.92. These differences in BC emission factors suggest that ≈ 2.5 times higher BC ship emissions might be more appropriate for future transit and oil-gas-related shipping than the original estimate from Peters et al. (2011) . However, note that McKuin and Campbell (2016) also point out that small fishing vessels (< 100 gross tonnage), which are not included in the analysis by Peters et al. (2011), contribute substantially to ship emissions. Neglecting these emissions from fishing activity likely leads to an underestimation of background ship emissions. This is important because higher background emissions might lead to a smaller impact of future transit and oil/gas related shipping (i.e. smaller relative increase of total aerosol emissions).

Tenfold ship emissions are achieved if we consider that i) transit shipping (which contributes most to the ship emissions by Peters et al. (2011) over the pristine Arctic Ocean between July and October) might be up to 4.6 times higher according to Corbett et al. (2010) and ii) the BC emission factor used by Peters et al. (2011) is likely underestimated by a factor of ≈ 2.5. Increasing the additional ship emissions (both transit shipping and oil/gas related shipping) from Peters et al. (2011) by a factor of ten is an upper estimate and is probably too high to represent conditions in 2050.

2.1.3 Heterogeneous freezing of mixed-phase clouds in ECHAM6-HAM2

In ECHAM6-HAM2, ~~BC particles~~ dust and BC particles (also those emitted by ships) can act as INPs in the immersion mode when transferred to the internally mixed mode. Heterogeneous freezing in ECHAM5-HAM is thoroughly described in the study of Hoose et al. (2008)~~, more equations can be found therein~~. The only differences in ECHAM6-HAM2 are that i) ~~only contact freezing by montmorillonite dust and immersion freezing by montmorillonite dust and BC are considered~~ contact freezing is limited to montmorillonite dust because contact freezing of BC is controversial and that ii) only particles in the accumulation and coarse ~~mode~~ modes can induce freezing. The freezing rate is defined as the number of cloud droplets that freeze per time and volume of air. Among other factors such as temperature, the contact freezing rate depends on the volume-mean droplet radius as well as the CDNC, while the immersion freezing rate depends on the cloud water mixing ratio.

2.1.4 Calculation of aerosol radiative forcings and cloud radiative effects

Both aerosol radiative forcings and CREs are calculated online by calling the radiation scheme once with and once without considering aerosol particles or clouds~~(all-sky and clear-sky conditions, respectively)~~; the difference between the two radiation calls is ~~considered as the instantaneous aerosol radiative forcingor CRE~~called radiative forcing due to aerosol-radiation interactions (RF_{ari}) for aerosols and CRE for clouds. While RF_{ari} is normally used for the forcing by anthropogenic emissions being the only external forcing to the system, a double radiation call with zero aerosols as the reference provides the sum of the natural and anthropogenic radiative forcing. For SW radiation, aerosol radiative forcings and CREs both depend on the surface

albedo. For example, an aerosol particle that scatters SW radiation can either have a cooling or a warming effect depending on whether the ~~underlaying~~ underlying surface has a lower or a higher surface albedo, respectively. Since the surface albedo decreases in our future simulations due to melting of sea ice, changes in ~~the radiative effects~~ RF_{ari} and CRE can either be caused by changes in aerosol/cloud properties or changes in surface albedo. For clouds, we can distinguish the two causes by applying the cloud radiative kernel method described in the study of Zelinka et al. (2012), which is independent of changes in surface albedo. With this method, we can furthermore disentangle changes in LW CRE caused by changes in clouds from those caused by ~~e.g.~~ surface temperature changes (see also Shell et al., 2008). A higher surface temperature enhances the outgoing LW radiation from the surface. Thus, more LW radiation can be absorbed by clouds and the LW CRE increases. In addition, the cloud radiative kernel method allows for diagnosis of how different cloud types (low and free-tropospheric clouds; Zelinka et al., 2016) and changes in different cloud properties (cloud cover/amount, cloud optical thickness, and cloud top altitude) contribute to the total changes in CREs. Note that with this method, gridboxes without incoming radiation are set to missing values for both SW and LW CRE. While this is not an issue for July, August, and September, most values between $85°$ and $90°$ N are missing in October. For the SW CRE, we set these missing values to zero; for the LW CRE, September values instead of the mean over September and October are shown for these gridboxes.

In our model, the reduction of snow albedo due to deposited BC is determined through interpolations of a lookup table based on a single-layer application of the SNICAR model (Flanner et al., 2007). The BC concentration in the top $2\,\mathrm{cm}$ of snow is considered. The concentration depends on the surface influx of snowfall as well as the influx of BC removed from the atmosphere through dry deposition, wet deposition, and sedimentation. Both BC scavenged by hydrometeors through in-cloud (Croft et al., 2010) and below-cloud ~~(Croft et al., 2010) wet deposition are~~ (Croft et al., 2009) wet deposition is assumed to reach the surface within one timestep (if ~~hyrometeors~~ hydrometeors do not evaporate in subsaturated regions below clouds). Given that both the spatial and the temporal ~~resolution~~ resolutions of our model are low ($1.875° \times 1.875°$; $7.5\,\mathrm{min}$), this assumption seems justified. The concentration of BC in snow can be further modified through scavenging by snow melt and glacier runoff. Since the scavenging ratios are low (0.2 for BC particles in the internally mixed mode and 0.03 for those in the externally mixed mode; Flanner et al., 2007), the BC concentration in snow increases after snow melt. Lastly, while albedo reductions of snow on land and on sea ice are considered, the impact of BC deposition on bare sea ice is not. This is due to the different characteristics of the sea ice albedo concerning its interaction with the deposited BC, which would only lie on top of the ice instead of being mixed-in. However, as the spatial coverage of bare sea ice without any snow cover is small in the model, the impact of omitting this darkening is expected to be negligible.

2.2 Model simulations

A summary of the model simulations can be found in Table 1. ECHAM6-HAM2 is an atmosphere-only model, i.e. SIC and SST need to be prescribed (see Section 2.3). To estimate the impact of future Arctic warming and sea ice retreat on aerosol particles and clouds, we conducted simulations under present-day (year 2004) and future (year 2050) conditions. The following simulations were performed with a resolution of T63L31 (corresponding to ~~$\approx 1.875° \times 1.875°$~~ $\approx 1.875° \times 1.875°$ with 31 vertical levels):

– **arctic_2004**: Global greenhouse gas concentrations, SIC, SST, and ~~ship emissions~~ prescribed aerosol emissions (including ships) from the year 2004 are used.

– **arctic_2050_EM2004**: The global greenhouse gas concentrations in the year 2050 follow IPCC's Representative Concentration Pathway RCP8.5 (Collins et al., 2013). To prescribe future SIC and SST, we used results from an Earth System Model (ESM; see Section 2.3) simulation. The same prescribed aerosol emissions are used as in 2004. Therefore, all anthropogenic aerosol emissions between **arctic_2004** and **arctic_2050_EM2004** are identical.

– **arctic_2050**: The same as **arctic_2050_EM2004** but the prescribed aerosol emissions are representative for ~~2050. The emission factor~~ 2050 (RCP8.5). The emission factors of SO_2 ~~ship emissions is~~ and OC ship emissions are smaller than in **arctic_2050_EM2004** since regulations and technological improvements are taken into account~~(see Sect. ??).~~ . Additional Arctic ship emissions are not accounted for.

– **arctic_2050_shipping**: The same as **arctic_2050** but with additional ship emissions in the Arctic. These emissions are estimated from ~~Peters et al. (2011, see Section ??)~~ Peters et al. (2011, see Section 2.1.2) based on future transport and oil/gas extraction. Since ~~the impact of~~ these additional Arctic ship emissions ~~was hardly noticeable~~ induced no significant changes in our test simulations (not shown), we increased the emissions by a factor of ten (mass flux). By comparing **arctic_2050** with **arctic_2050_shipping**, we can estimate the impact of future Arctic ship emissions enabled by the smaller SIC.

Each simulation is run for ~~10~~ 20 years with the same forcing for each year, therefore yielding ~~10~~ 20 ensemble members.

2.3 ~~Ship emission inventory~~

~~We used the ship emission inventory by Peters et al. (2011) for both present-day and future global ship emissions. It includes the species SO_2, BC, and OC for the years 2004, 2030, and 2050. For this study, we only use emissions from 2004 and 2050. The shipping emissions for the year 2004 are based on Dalsøren et al. (2009) , who combined the observational data sets COADS and AMVER. Considering only ships above 100 gross tons, Dalsøren et al. (2009) found a total fuel consumption of for the year 2004.~~

~~Peters et al. (2011) used this 2004-inventory also as a "background" for the years 2030 and 2050. They assumed that emission factors of SO_2 will decrease due to regulations and improved technology but that everything else (other aerosol emission factors ; shipping routes) will remain constant. In addition, Peters et al. (2011) calculated Arctic ship emissions for the years 2030 and 2050 for transit shipping and for shipping that is related to oil and gas production. Changes in ship emissions from the sectors tourism, fishery, and local/national transport are not considered. For the year 2004, no transit shipping was assumed, and the oil and gas shipping was estimated based on oil tankers operating in the Arctic region. The expected increase in these two sectors is related to SIC: less sea ice will faciliate the passage through the Arctic ocean and exploit new areas to oil and gas production. Peters et al. (2011) used different models to create their ship emission inventory; for more details we refer to their study. We processed the data from Peters et al. (2011) to create input files that are compatible with our model.~~

Table 1. An overview of the different model simulations.

Model simulation	Greenhouse gas concentrations	SIC/SST	~~Anthropogenic~~ Ship emissions	Other anthropogenic aerosol emissions
arctic_2004	Year 2004	Year 2004 (AMIP)	Year 2004 (Dalsøren et al., 2009)	Year 2004 (ACCMIP)
arctic_2050_EM2004	Year 2050 (RCP8.5)	Year 2050 (MPI-ESM RCP8.5)	Year 2004 (Dalsøren et al., 2009)	Year 2004 (ACCMIP)
arctic_2050	Year 2050 (RCP8.5)	Year 2050 (MPI-ESM RCP8.5)	Dalsøren et al. (2009) with emission factors for 2050	Year 2050 (ACCMIP RCP8.5)
arctic_2050_shipping	Year 2050 (RCP8.5)	Year 2050 (MPI-ESM RCP8.5)	~~Year~~ Dalsøren et al. (2009) with emission factors for 2050 ~~+~~ and additional ship emissions by Peters et al. (2011)	Year 2050 (ACCMIP RCP8.5)

~~As mentioned previously, we increased ship emissions by a factor of ten to detect a noticeable signal in aerosol particles. This is in agreement with the results of Peters et al. (2014), who studied the effect of ship emissions on tropical warm clouds with ECHAM5-HAM. In the following, we explain why we consider the tenfold higher emissions to be an upper estimate of future Arctic ship emissions.~~

~~Recently, McKuin and Campbell (2016) pointed out that both global and Arctic ship emission inventories might underestimate BC ship emissions because small fishing vessels (< 100 Gt) were not included in the analysis and because too low BC emission factors were used. In this study, we do not consider future changes in fishing activity. Therefore, both present-day and future background ship emissions should be underestimated by the same absolute amount in our analysis. If these background ship emissions were to occur over the Arctic Ocean or its vicinity, such an increase in background emissions might lead to a smaller impact of future transit and oil/gas related shipping (i.e. smaller relative increase of total aerosol emissions). This is an argument against increasing the Arctic ship emissions. A higher BC emission factor, on the other hand, would increase ship emissions from all sectors, although not necessarily by the same coefficient since BC ship emissions depend on engine and fuel type. While the ship emission inventory by Peters et al. (2011) used a BC emission factor of 0.35, McKuin and Campbell (2016) found – depending on the averaging method and the area (non-emission control versus emission control) – factors between 0.79 and 0.92. This suggests that $\approx$ 2.5 times higher BC ship emissions might be more appropriate for future transit and oil-gas-related shipping than the original estimate from Peters et al. (2011). Again, increases in background shipping emissions need to be considered as well.~~

~~Compared with other estimates of future Arctic transit shipping, the results from Peters et al. (2011) lie between those from Paxian et al. (2010) and Corbett et al. (2010): the fuel consumption by Paxian et al. (2010) is 1.4 to 2.4 times smaller than the values reported by Peters et al. (2011) . Depending on the scenario, the estimated CO_2 emissions by Corbett et al. (2010) are 2 to 4.6 times higher in 2050 than the values reported by Peters et al. (2011).~~

~~Averaged over the period from July to October, transit shipping contributes more than petroleum-related shipping in the study from Peters et al. (2011), especially over the central Arctic Ocean. For transit shipping, the impact of ship emissions on climate is thus larger than for petroleum-related shipping since considerably higher emissions occur in regions where the background aerosol concentrations are lower.~~

~~Considering now that i) the BC emission factor used by Peters et al. (2011) is likely underestimated by a factor of $\approx$ 2.5, ii) transit shipping might be up to 4.6 times higher according to Corbett et al. (2010), and iii) transit shipping contributes most to the ship emissions over the aerosol-poor Arctic Ocean, increasing the additional ship emissions (both transit shipping and oil/gas related shipping) from Peters et al. (2011) by a factor of ten can be considered as realistic, tough upper estimate. Furthermore, even if the tenfold higher ship emissions are probably too high to represent conditions in 2050, such high emissions could be reached in later years when population growth might lead to an increase in trade and petroleum demand.~~

2.3 Boundary conditions

Both SIC and SST are prescribed in ECHAM6-HAM2. For future conditions, we used model results from the Earth System Model MPI-ESM as input (simulation for the climate model intercomparison project phase 5 (CMIP5), RCP8.5; Giorgetta et al., 2013) . We chose MPI-ESM because its atmospheric component is ECHAM and ~~because~~ the simulated future sea ice retreat is close to the model median of CMIP5. ~~A drawback of our study~~ An inconsistency in our simulations is that we did not apply the SST and SIC mid-month correction to the MPI-ESM data as recommended by Taylor et al. (2000), which is applied for the AMIP data that we used for the year 2004 (Taylor et al., 2000). Therefore, the seasonal variability in SIC and SST is somewhat underestimated ~~.~~ in 2050. However, compared to the large differences in SIC and SST between 2004 and 2050, we do not expect this to affect the main conclusions of our study.

As mentioned previously, future greenhouse gas emissions follow the RCP8.5 scenario, which shows a similar CO_2 emission increase as the A2 scenario that Peters et al. (2011) assumed in their analysis. From 2004 to 2050, the global greenhouse gas volume mixing ratios change as follows: from ~~377~~ 377 ppm to 541 ppm for CO_2, from ~~1.76~~ 1.76 ppm to 2.74 ppm for CH_4, from ~~319~~ 319 ppb to 367 ppb for N_2O, from ~~256~~ 256 ppt to 107 ppt for CFC-11, and from ~~540~~ 540 ppt to 345 ppt for CFC-12 (CFCs are chlorofluorocarbons). Also most prescribed aerosol emissions (excluding DMS terrestrial emissions, biogenic organic carbon emissions, and ship emissions) follow RCP8.5, which decline in most industrial sectors from 2004 to 2050.

~~Since~~ We refrained from averaging SIC and SST ~~vary from year to year,~~ over several years (e.g. 2000-2010) to avoid having spurious regions with intermediate SIC and SST. However, the interannual variability in SIC is pronounced, therefore we performed test simulations using SIC and SST from: i) the ~~year~~ years 2003 and 2004 from AMIP and ii) the first and the second ensemble ~~member~~ members from the MPI-ESM CMIP5 simulation for the year 2050. Overall, the Arctic SIC in 2003 was somewhat smaller than in 2004, and the SIC in the first ensemble member from MPI-ESM was smaller than in the second

ensemble member. ~~However, we~~ We found that the basic results and main conclusions do not depend on these differences in SIC but looking at only two years for both present-day and future might not be sufficient to confirm that all our results are robust. In the following, we will ~~therefore~~ always refer to the simulations using SIC and SST from 2004 and future SIC and SST from the first ensemble member of MPI-ESM.

To verify consistency between future shipping routes and sea ice extent, we further compared the sea ice conditions used to calculate future ship emissions with the sea ice conditions employed in our simulations (Appendix B).

2.4 Statistical test

Wilks (2016) recently pointed out that the ~~"naive stippling approach ",~~ approach to accept alternative hypotheses at any gridpoint where locally significant results occur (which is commonly used in atmospheric sciences~~,~~) leads to overstatements of scientific results: with ~~the~~ this so-called "naive stippling approach", a significance test is calculated for every gridpoint and all gridboxes are stippled where the p-value is smaller than $5\,\%$ (for a significance level of $\alpha = 0.05$). This approach has two main limitations: 1. Assuming that the spatial correlation is zero, $5\,\%$ of the gridboxes show on average stippling *just by chance*. 2. Spatial autocorrelation – often large when analysing gridded climate data – increases the false discovery rate (FDR) for the "naive stippling approach", i.e. the null hypothesis is often rejected although it is true. As suggested by Wilks (2016), we circumvent the problem by controlling the FDR instead. The advantages of this approach are the elimination of many spurious signals and the robustness concerning spatial correlation. In this method, a threshold p-value is calculated below which the result is supposed to be signal, not noise. We assume that the spatial correlation is moderate or large for the variables we are looking at. Therefore, we set α_{FDR} to ~~$2 \times \alpha$~~ $2 \cdot \alpha$ (see Wilks, 2016, for explanation). For the individual gridpoints, p-values are calculated using the Wilcoxon-Mann-Whitney test instead of the often used Welch's test since the latter is only valid if the samples are normally distributed (a condition which was sometimes not met in our results). The only exception where we used the Welch's test is for testing the significance of the results from the cloud radiative kernel method (see Appendix C): we could not apply the Wilcoxon-Mann-Whitney test to the cloud radiative kernel results because they are given as differences instead of absolute values. Throughout this paper, the term "significant" is interchangeable with "statistically significant".

3 Results and Discussion

First, the changes in natural aerosol populations, clouds, and their radiative forcings/effects in a warming Arctic ~~will be~~ are assessed (Sect. 3.1). Second, we ~~will~~ determine the influence of additional Arctic shipping activity related to transit shipping and petroleum activites on climate (Sect. 3.2).

Most figures ~~will~~ show the mean over the ~~ten~~ twenty ensemble members for the reference simulation on the left and differences between the perturbed ensemble mean and the reference ensemble mean on the right. As mentioned previously, we ~~will~~ analyse the months July to October. Since the conditions ~~(e.g. SW radiation, temperature) considerably change~~ change considerably from July to October, averaging over these four months might hide significant changes occurring in only one or two months. Therefore, we decided to average the results from July to August (late summer) and from September to October

(early autumn). If the season is not specified in the text, results refer to both late summer and early autumn. Most of the figures show results for early autumn, except for changes in clouds and RF_{ari} associated with enhanced Arctic shipping, which refer to late summer. When we compare our results to other studies, we average over the same time and area as the authors of the corresponding study did for a meaningful comparison.

Each simulation consists of ~~ten~~ twenty ensemble members to account for the high variability in Arctic climate. However, uncertainties associated with the used climate model can of course not be captured with this approach. It is well known that different global climate models ~~considerably deviate~~ deviate considerably, e.g. when simulating aerosol-cloud interactions. Furthermore, ~~most models~~ models of different resolutions generally have problems to reproduce the structure of mixed-phase clouds prevalent in the Arctic (Morrison et al., 2009; Klein et al., 2009; Fan et al., 2011; Morrison et al., 2011; Possner et al., 2017), and the future sea ice extent as well as the prescribed aerosol emissions are highly uncertain (Collins et al., 2013). To gain a better understanding of the robustness of our results, we ~~will~~ compare them with other studies, both concerning relative and absolute changes. In addition, we provide in the Supplementary Information a comparison of the simulation **arctic_2004** with Arctic observations. While the simulated ice water path (IWP) and the aerosol optical thickness (AOT; at least in some Arctic regions) have a low bias, the surface concentrations of BC and sulphate, the liquid water path (LWP), the cloud cover, and the SW, LW, and net CREs at the surface and the TOA agree well with the observations.

3.1 Changes due to warming and sea ice retreat

In the following, we ~~will analyse how~~ analyse how a future temperature increase in the Arctic affects natural aerosol particles, clouds, and radiation. For that, ~~the~~ simulation **arctic_2050_EM2004** ~~will be~~ is compared with **arctic_2004**. The Arctic sea ice area decreases from $6.1 \cdot 10^6$ km^2 to $3.4 \cdot 10^6$ km^2 and from $5.7 \cdot 10^6$ km^2 to $2.3 \cdot 10^6$ km^2 in late summer and early autumn, respectively. To gain some insight into the importance of this retreat in sea ice, we averaged some vertically integrated variables such as AOT or CDNC burden over four different regions north of 60° N (see Tables 2 and 3 for late summer and early autumn, respectively): i) the whole region north of 60° N, ii) regions with open ocean in both 2004 and 2050 (SIC < 0.5), iii) regions with sea ice coverage in both 2004 and 2050 (SIC > 0.5), and iv) regions that are covered with sea ice in 2004 (SIC > 0.5), but not anymore in 2050 (SIC < 0.5). This analysis is only qualitative since advection can hide significant changes related to the sea ice retreat, the SIC values used for the calculations are monthly means, and the threshold of 0.5 for SIC to differentiate open ocean and sea ice is somewhat arbitrary.

3.1.1 Aerosol particles

~~In the vicinity of the Arctic Ocean, both DMS and sea salt burdens significantly increase (not shown), which is predominantely caused by~~ Over the central Arctic Ocean, the decrease in SIC (Fig. 2) enables emission fluxes of DMS and sea salt, which significantly increase their burdens (Supplementary Fig. 4; Tables 2, 3). As a second-order effect, significant increases in u_{10} (Supplementary Fig. 5) over the central Arctic Ocean in early autumn increase sea salt and DMS emissions. In regions where the SIC does not change, both changes in u_{10} (insignificant) and changes in SST (Supplementary Fig. 6) affect DMS and sea

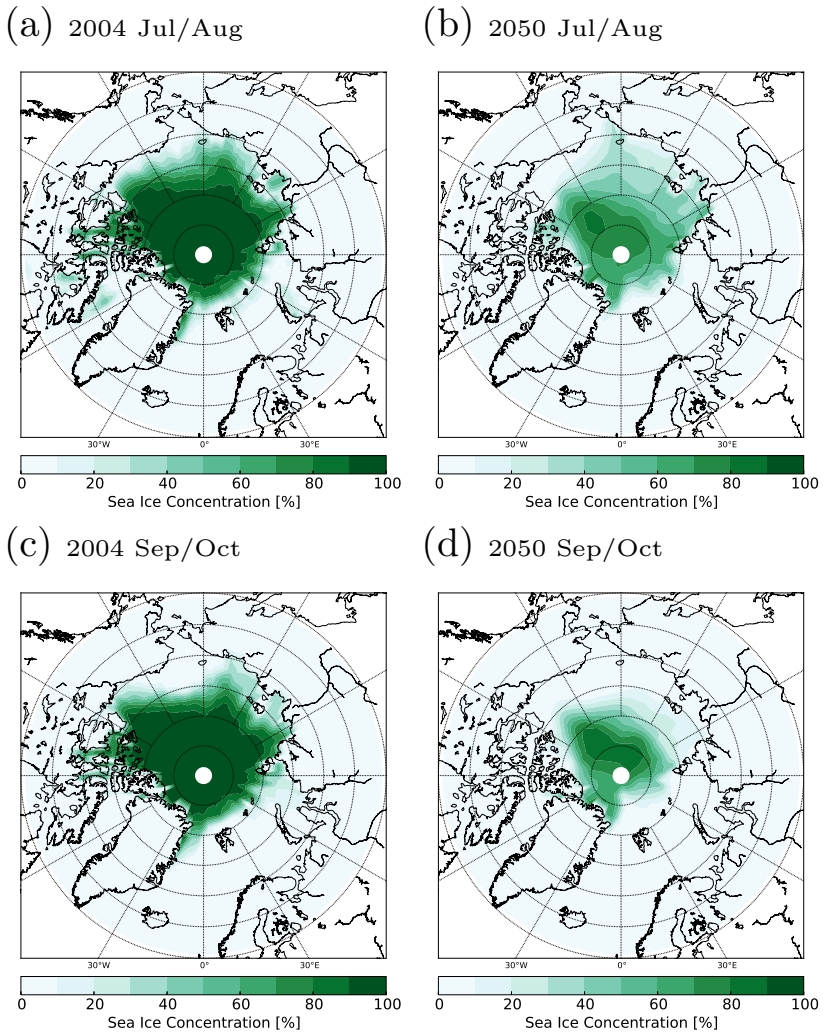

Figure 2. SIC in 2004 and 2050 for late summer (Jul/Aug) and for early autumn (Sep/Oct).

salt emissions, and thus their burdens. For example, the decrease in the sea salt burden over the Bering Strait is due to decreases in SST (caused by a model bias in the MPI-ESM sea surface temperature compared to AMIP) and u_{10}.

Despite the pronounced increases in DMS burden, the sulphate burden ~~does not change significantly~~ shows no large changes since it is dominated by other emissions (e.g. anthropogenic SO_2 emissions; not shown). Also the aerosol size distributions at 950 hPa (corresponding to $\approx$ 540 m; Fig. 3a) and 800 hPa (corresponding to $\approx$ 1950 m; Fig. 3b) show only small, non-significant changes from 2004 to 2050 (shown for early autumn; averaged between ~~70°~~ 75° and 90° N). The number concen-

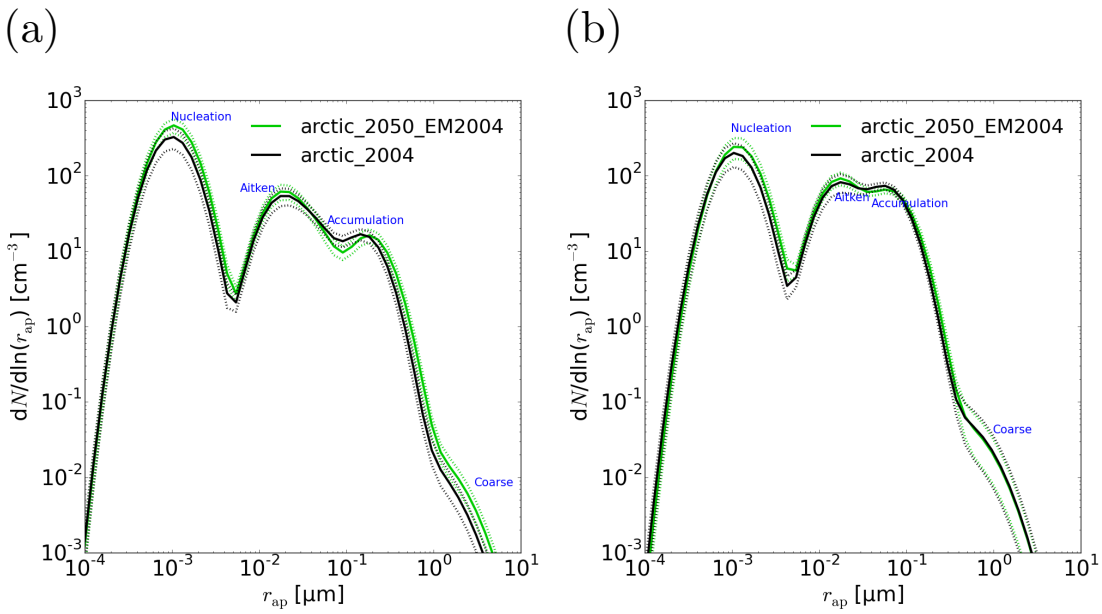

Figure 3. Aerosol number size distributions in 2004 (**arctic_2004**) and 2050 (**arctic_2050_EM2004**); N stands for the number concentration (assuming that ~~$1\,kg_{air} \approx 1\,m^3$~~ air density $\rho_{air} \approx 1\,kg\,m^3$), r_{ap} for the radius of the aerosol particles. The size distributions are shown for early autumn (Sep/Oct) at 950 hPa (a) and 800 hPa (b), averaged between 75° and 90° N. The solid lines denote ensemble means, the dotted lines the subtracted/added standard deviations. Different colors (black, green) stand for different simulations (see legend).

tration slightly increases in ~~both the nucleation and the coarse mode . We attribute the enhanced number concentrations in the coarse mode mainly to direct sea salt emissions and the increase in the nucleation mode to DMS emissions; the latter~~ the nucleation mode in both seasons, which we attribute to the enhanced DMS emissions. DMS is oxidised via SO_2 to sulphuric acid, which can form new particles. In late summer, the number concentration in the Aitken mode increases to some extent. In early autumn, the number concentration decreases at ~~$r_{ap} \approx 0.1\,\mu m$~~ $r_{ap} \approx 0.1\,\mu m$ (r_{ap} is the radius of the aerosol particles), which might be caused by ~~(non-significant)decreases in~~ decreases in BC and OC burdens (not shown), but increases in the coarse mode. The smaller BC and OC burdens ~~.~~ can be explained by the increase in precipitation, which leads to enhanced wet deposition (the BC and OC emissions are identical between the two simulations). The increased number in the coarse mode can be explained by the increase in sea salt emissions.

Struthers et al. (2011) compared sea salt emissions for a nearly ice-free summer (2100) with present-day conditions (2000) and found an increase in mass emissions ~~from 7.1 $\mu g m^{-2} s^{-1}$ to 30.5 $\mu g m^{-2} s^{-1}$ (factor of ≈ 4);~~ by a factor of ≈ 4 (present-day value 7.1 $\mu g\,m^{-2}\,s^{-1}$); this is an average over JJA (June, July, August) and ~~70~~ 70° to 90° N. Note that we chose 2050 for our simulations due to the availability of Arctic ship emissions for this year. In the same region, Browse et al. (2014) found

Table 2. Absolute values for the year 2004 and differences between 2050 and 2004 (i.e. between simulations **arctic_2050_EM2004** and **arctic_2004**) for sea salt burden, DMS burden, AOT, LWP, IWP, cloud cover ("CC"), in-cloud CDNC burden, and T_{surf} for late summer (Jul/Aug). The numbers are averaged over four regions between 60° and 90° N: i) the whole region, ii) gridboxes which are ocean in both 2004 and 2050 (SIC < 0.5; "Ocean"), iii) gridboxes which are covered by sea ice in both 2004 and 2050 (SIC > 0.5; "Sea ice"), and iv) gridboxes which have sea ice in 2004 (SIC > 0.5) but not in 2050 (SIC < 0.5; "Transition"). Significant changes are marked with a star. Note that the SST is prescribed, i.e. shows no interannual variability.

	Total region		Ocean		Sea ice		Transition	
	2004	2050 – 2004	2004	2050 – 2004	2004	2050 – 2004	2004	2050 – 2004
Sea salt (10^{-7} kg m^{-2})	1.2	0.18*	3.0	0.36*	0.18	0.12*	0.29	0.28*
DMS (10^{-7} kg m^{-2})	1.5	0.27*	3.2	0.39*	0.66	0.42*	0.92	0.73*
AOT (10^{-2})	3.6	0.26*	3.9	0.19	1.3	0.19*	1.6	0.19*
LWP (g m^{-2})	96	8.0*	108	7.3*	65	5.1*	67	7.9*
IWP (g m^{-2})	17	0.00	15	0.00	12	0.09	14	-0.06
CC (%)	77	0.08	81	0.85	88	-0.35	82	0.50
CDNC (10^{10} m^{-2})	6.0	0.47*	5.1	0.30*	1.9	0.22*	2.4	0.34*
T_{surf} (K)	281	0.98*	278	1.6*	273	-0.36*	272	0.23*

Table 3. As Table 2 but for early autumn (Sep/Oct).

	Total region		Ocean		Sea ice		Transition	
	2004	2050 – 2004	2004	2050 – 2004	2004	2050 – 2004	2004	2050 – 2004
Sea salt (10^{-7} kg m^{-2})	2.7	0.84*	6.6	2.0*	0.29	0.27*	0.45	0.53*
DMS (10^{-7} kg m^{-2})	0.62	0.12*	1.1	0.18*	0.32	0.24*	0.47	0.34*
AOT (10^{-2})	3.2	0.28*	3.5	0.32	1.3	0.03	1.4	0.21*
LWP (g m^{-2})	72	5.3*	92	2.0	24	14*	37	19*
IWP (g m^{-2})	21	0.59*	21	0.79*	12	0.17	14	0.76
CC (%)	87	0.05	89	-0.70*	92	1.3*	92	2.3*
CDNC (10^{10} m^{-2})	4.0	0.31*	4.3	0.30	0.96	0.28*	1.3	0.47*
T_{surf} (K)	271	2.8*	277	1.8*	258	7.9*	264	7.4*

that sea salt emissions increased by a factor of 10 ~~to $\approx 0.069\,\mu g m^{-2} s^{-1}$~~ (present-day value $6.9 \cdot 10^{-3}\,\mu g\,m^{-2}\,s^{-1}$) in August when comparing a hypothetically ice-free ocean with present-day conditions (2000). In our simulations (70° to 90° N), sea salt emissions increase ~~from $1.48 \times 10^{-3}/2.43 \times 10^{-3}\,\mu g m^{-2} s^{-1}$ (JJA/August) to $2.79 \times 10^{-3}/4.00 \times 10^{-3}\,\mu g m^{-2} s^{-1}$ from 70-90° N in 2050, i.e.~~ by a factor of ~~< 2. As expected, the~~ 1.8 and 1.7 in JJA and August by 2050, respectively, compared to the present-day values of $1.52 \cdot 10^{-3}$ and $2.42 \cdot 10^{-3}\,\mu g\,m^{-2}\,s^{-1}$. The relative increase in emissions is largest in the study by Browse et al. (2014), where the absolute decrease in SIC is largest, and smallest in our study, where the absolute decrease in SIC is smallest. Present-day emissions are a factor of ~~≈ 3~~ ≈ 3 lower in our simulations compared with Browse et al. (2014), which results from the differences in the two parameterisations ~~(modified Long et al., 2011; Gong, 2003)~~ (Gong, 2003; Long et al., 2011, with SST corrections) as shown in the study of Long et al. (2011). The absolute present-day emissions reported by Struthers et al. (2011) are at least three orders of magnitudes higher than in our simulations. This might again be caused by the parameterisations used since differences in u_{10} and SST are too small to explain the large disagreement. Struthers et al. (2011) used a modification of the Mårtensson parameterisation combined with the Monahan parameterisation for particles $> 1.4\,\mu m$ (Mårtensson et al., 2003; Monahan et al., 1986). However, neither Long et al. (2011) using the Mårtensson parameterisation nor us using the Monahan parameterisation for particles $r_{dry} < 4\,\mu m$ (in earlier simulations with ECHAM-HAM; not shown) found ~~as high emissions~~ emissions as high as Struthers et al. (2011). Therefore, we expect that

differences in the number fluxes of large particles ($> 4\,\mu m$), which contribute most to mass emissions (Long et al., 2011), are responsible for the large discrepancy. Note that at the present state, sea salt emissions thus remain highly uncertain and differ considerably between models: a comparison of 11 chemical transport and global climate models showed that the global annual mass emissions of sea salt lie in the range between $2.2 \cdot 10^{12}$ and $9.9 \cdot 10^{12}\,kg\,yr^{-1}$ for nine of the models; two models calculate higher emissions of $22 \cdot 10^{12}\,kg\,yr^{-1}$ and $118 \cdot 10^{12}\,kg\,yr^{-1}$ (de Leeuw et al., 2011). For comparison, our simulated value is lower than in all of these models ($1.3 \cdot 10^{12}\,kg\,yr^{-1}$) because our parameterisation does not account for the contributions from spume drops (Long et al., 2011), but is on the same order of magnitude as most of them. Our simulated absolute increases in sea salt mass emissions might therefore be underestimated.

3.1.2 Clouds

Except for cloud cover, LWP, and IWP, the averages of cloud properties (such as ~~liquid water content~~ LWC or CDNCs) refer to in-cloud values, i.e. by averaging only over periods and locations when and where clouds are present.

In general, the number of aerosol particles acting as CCN increases in the future, which leads to enhanced CDNCs (~~shown in~~ Fig. 4d~~for early autumn). Also the liquid water content~~). The increase in the number of CCN is not only caused by the increases in oceanic aerosol emissions but also by changes in meteorology: the updrafts available for activation increase in the boundary layer between 75° and 90° N in early autumn (Supplementary Fig. 7), which supports the formation of cloud droplets in this region. Averaged between 75° and 90° N, the CDNC burden increases by 10% and 29% in late summer and early autumn, respectively. Relative changes are largest in regions where sea ice melted (Tables 2, 3). Also LWC increases (see Fig. 4b) because both the open ocean and higher air temperatures increase the specific humidity. The increase in ~~liquid water content~~ LWC can be ascribed to both higher CDNCs and larger cloud droplets (not shown). Averaged between 75° and 90° N, LWP increases by 10% in late summer and by 34% in early autumn. Precipitation shows ~~no significant changes.~~ significant increases in early autumn (Supplementary Fig. 8). In late summer, changes are only significant when averaged between 60° and 90° N and smaller than in early autumn (+4% compared to +9%).

We also obtain ~~increases~~ increased CDNCs (which we attribute to ~~increases in~~ increased CCN concentrations) when averaging over ~~cloudy and non-cloudy~~ all-sky conditions. In contrast, Browse et al. (2014) found small decreases in CCN concentrations (~~averaged over cloudy and non-cloudy~~ also averaged over all-sky conditions) over the Arctic Ocean. In their simulations, the liquid clouds over the ocean suppressed new particle formation via aqueous phase oxidation of SO_2 (a process also considered in ECHAM6-HAM2). Instead, particles grew to larger sizes and were efficiently scavenged by drizzle. The different responses when compared to our simulations could e.g. be caused by different oxidant concentrations (H_2O_2, O_3) or by the different handling of drizzle and precipitation: Browse et al. (2014) derived drizzle rates from Arctic observations of cloud altitude and droplet concentrations and scaled them by the low-cloud fraction. On the other hand, cloud microphysical processes (e.g. diffusional growth, coagulation) are explicitly calculated in our simulations and coupled with aerosol particles via Köhler theory and freezing parameterisations. Drizzle is not considered as a separate size class in our simulations; however, Sant et al. (2015) ~~have shown with ECHAM5-HAM~~ showed that the impact of drizzle on the CDNC burden is rather small in the Arctic in ECHAM5-HAM.

As expected, the higher temperatures in 2050 influence the occurrence of cloud ice (both cirrus and mixed-phase) in our simulations by shifting the isotherms and thus also cloud ice towards higher altitudes. Changes in ice water content (IWC) (Fig. 5b) can be caused by changes in the ICNC (Fig. 5d) and/or the effective ice crystal radius (Fig. 5f). ~~At altitudes below , both~~ Both changes in the ICNC and radius have ~~an influence~~ a considerable influence at altitudes below 500 hPa, whereas changes in radius dominate at higher altitudes. The increase of ICNC near the surface is mainly caused by enhanced convection, which leads to small but numerous ice crystals following the temperature-dependent empirical parameterisation of Boudala et al. (2002).

Compared to the pronounced increases in LWP, changes in the IWP are small and only significant over the whole Arctic region and over the ocean in early autumn (slight increases; see Tables 2, 3). This can be explained by ~~the increase in CDNC. Between and , the enhanced ice water content is linked to the increase in ice crystal radius.~~ two opposing effects: on the one hand, the total water path increases due to the higher specific humidity. On the other hand, the temperature increase leads to a higher fraction of liquid water to the total water path. In our simulatons, the first effect slightly dominates in early autumn.

Especially in early autumn, significant changes in cloud cover occur (see Fig. 6). Cloud cover decreases where convective precipitation is most enhanced (~~near Svalbard~~e.g. near Svalbard; see Supplementary Fig. 9) but increases where sea ice vanished, e.g. over the East Siberian Sea and the Beaufort Sea ~~where sea ice has vanished~~ (Fig. A1 shows a map of the Arctic Ocean where the regional seas are labelled). When averaged over the open ocean area, cloud cover shows rather small but significant decreases in early autumn, whereas it increases significantly and pronouncedly where sea ice melted (Table 3). The latter is consistent with the findings from Abe et al. (2016), who found increases in the October cloud cover caused by sea ice reduction~~(~~, which leads to an enhanced moisture flux to the atmosphere. Also in our simulations, the surface fluxes increase significantly over regions where sea ice melted (not shown).

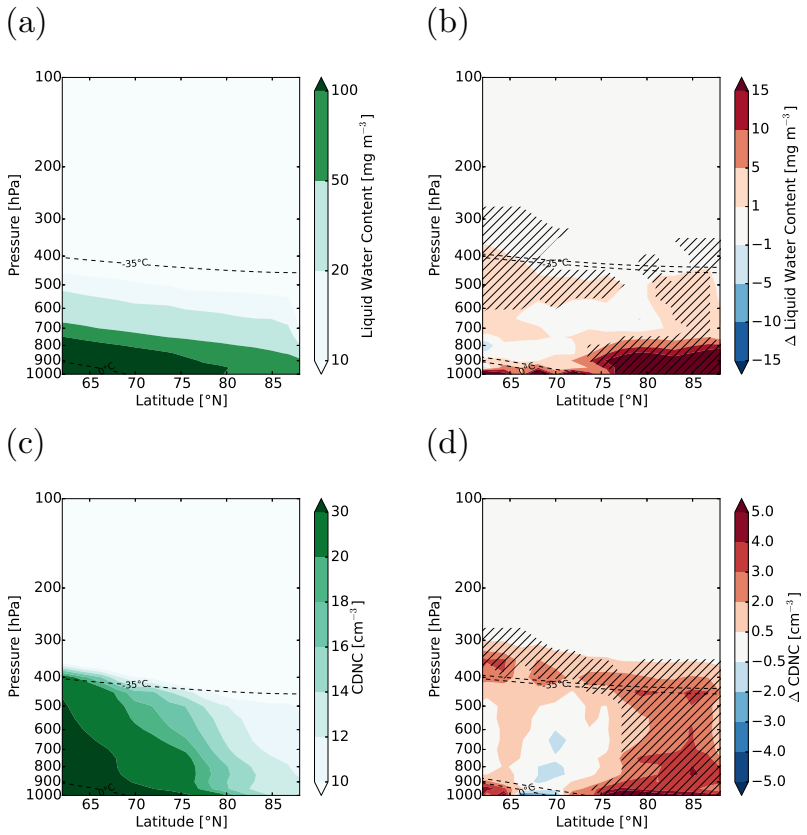

Figure 4. ~~Liquid water content~~ LWC and CDNC in 2004 in (a)/(c) and differences between 2050 and 2004 (i.e. between simulations **arctic_2050_EM2004** and **arctic_2004**) in (b)/(d) (~~all when/where clouds are present~~in-cloud values) in early autumn (Sep/Oct). ~~Stippled~~ Hatched areas are significant at the 95% confidence level. The dashed lines show the 0°C ~~-~~and the −35°C ~~-isotherms (for both 2004 and 2050 for the difference plots)~~isotherms.

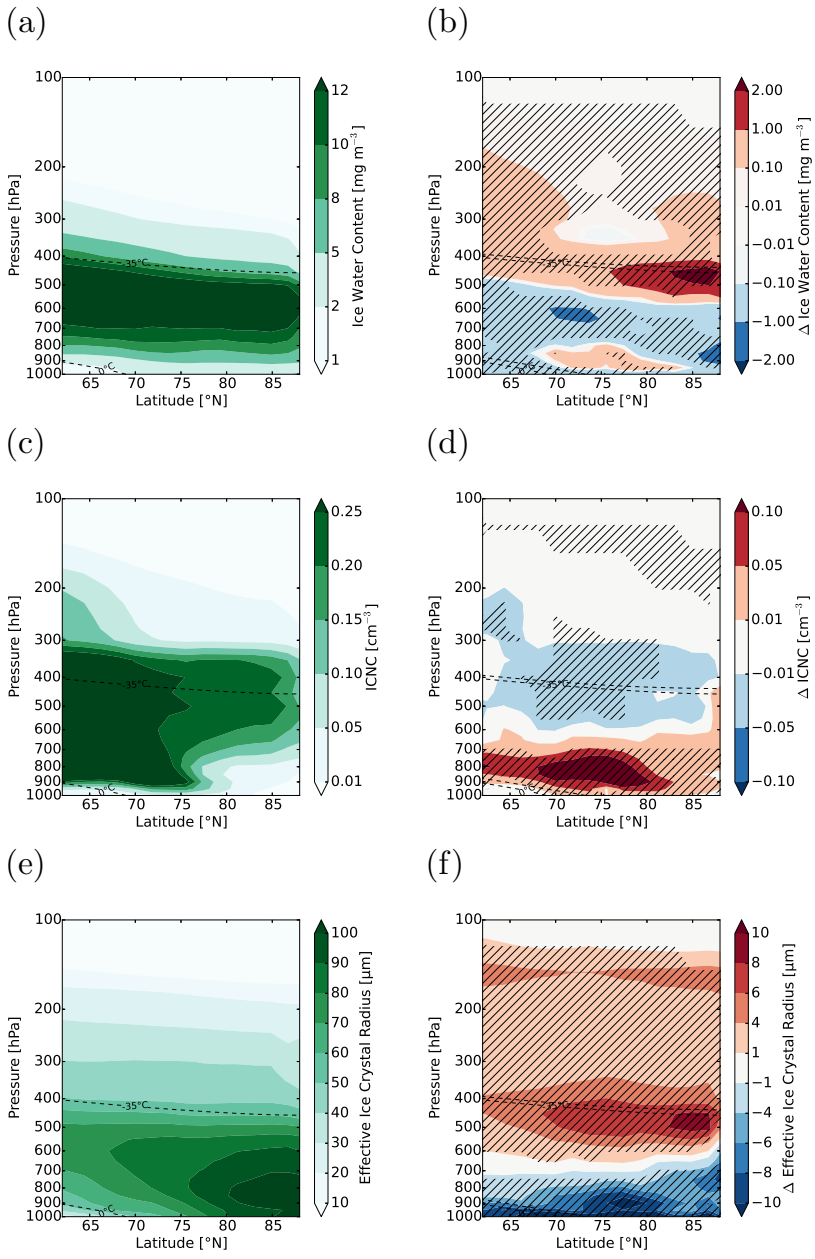

Figure 5. ~~Ice water content~~IWC, ICNC, and effective ice crystal radius in 2004 in (a)/(c)/(e) and differences between 2050 and 2004 (i.e. between simulations **arctic_2050_EM2004** and **arctic_2004**) in (b)/(d)/(f) (~~all when/where clouds are present~~in-cloud values) in early autumn (Sep/Oct). ~~Stippled~~ Hatched areas are significant at the 95% confidence level. The dashed lines show the 0°C ~~-~~and the -35°C ~~-isotherms~~isotherms. Note that they are zonally and temporally averaged, hence ice can exist at altitudes below the 0°C ~~-isotherm~~isotherm.

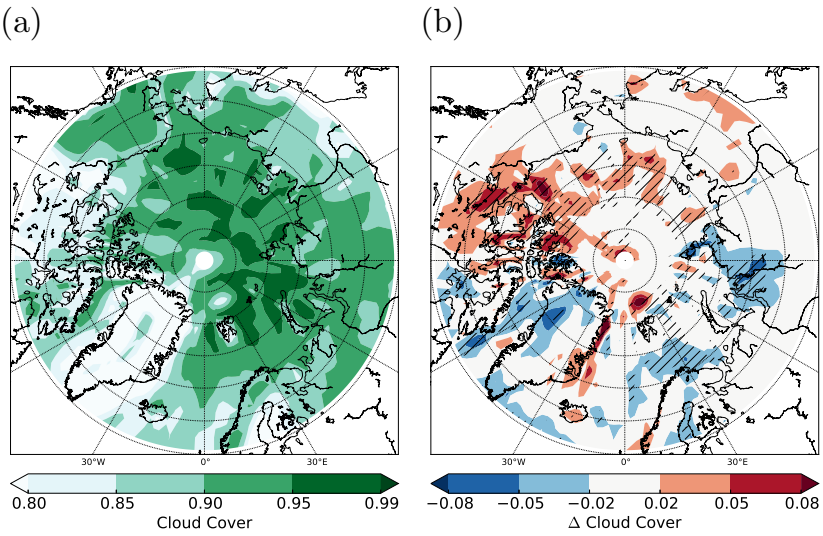

Figure 6. (a) Cloud cover in 2004 and (b) differences between 2050 and 2004 (i.e. between simulations **arctic_2050_EM2004** and **arctic_2004**) in early autumn (Sep/Oct). ~~Stippled~~ Hatched areas are significant at the 95% confidence level.

3.1.3 Aerosol radiative forcings

Unless otherwise stated, all aerosol radiative forcings and cloud radiative effects refer to those at the top of the atmosphere (TOA). As mentioned previously, RF_{ari} refers to the instantaneous effect of all aerosols on radiation. In 2004, aerosol particles have a negative RF_{ari} and thus cool the Arctic under clear-sky conditions (i.e. absence of clouds; see Fig. 7c), except over ~~the Arctic Ocean~~ sea ice and Greenland, where the surface albedo is high (see Fig. 7~~c~~a). If the presence of clouds is considered, aerosol particles warm the atmosphere also over Alaska and northeast Siberia (late summer) and over the whole northern Russia (early autumn; shown in Fig. 7e). Part of this warming might be caused by ~~absorbing aerosol layers lying~~ BC and dust aerosols above clouds (~~not shown): since~~ Supplementary Fig. 10): the clouds reflect more SW radiation than the snow/ice-free surface ~~,~~ ~~more~~ and part of the scattered SW radiation can also be absorbed by aerosol particles causing an increase in aerosol absorption as compared to clear-sky conditions (see e.g. Myhre et al., 1998). Moreover, the scattering of aerosol particles could become less important in the presence of clouds, which increases the relative importance of aerosol absorption to extinction. Averaged over the whole Arctic region, aerosol particles have a cooling effect under clear-sky conditions in 2004 ($-1.23\,\mathrm{W\,m^{-2}}$ for late summer and $-0.65\,\mathrm{W\,m^{-2}}$ for early autumn) but a warming effect if clouds are considered ($0.12\,\mathrm{W\,m^{-2}}$ for late summer and $0.09\,\mathrm{W\,m^{-2}}$ for early autumn). Note that changes at the surface are of opposite sign, i.e. the aerosol particles cool the surface under all-sky conditions. In our simulations, both the cooling and the warming are more pronounced in late summer than in early autumn due to the higher ~~availability of SW radiation~~solar zenith angle in late summer.

~~Increases in~~ Increases in the DMS and sea salt burdens ~~lead to significant increases in aerosol optical thickness (AOT)~~ increase the AOT in 2050 ~~, which are expected to induce a cooling since sea salt and sulphate particles are nearly pure scatterers. We cannot disentangle the aerosol radiative forcing induced by surface albedo changes from that induced by AOT changes. However, comparing aerosol radiative forcings~~ (significant changes from $1.6 \cdot 10^{-2}$ to $1.8 \cdot 10^{-2}$ in late summer and from $1.5 \cdot 10^{-2}$ to $1.7 \cdot 10^{-2}$ in early autumn; averaged over 75°-90° N). While the AOT does not significantly change over open ocean, it significantly increases over regions where ~~the surface albedo has not decreased – e.g. over the Norwegian Sea, the Greenland Sea, and the Barents Sea – shows that the increases in natural aerosol particles indeed lead to a significant cooling independent of surface albedo changes~~ sea ice melted (Tables 2, 3). The absorption aerosol optical thickness significantly decreases in early autumn ($1.16 \cdot 10^{-3}$ to $1.05 \cdot 10^{-3}$, averaged over 75°-90° N), which can be explained by the decrease in BC burden. In both late summer and early autumn, RF_{ari} shows significant decreases under both clear-sky (Fig. 7d; shown for early autumn) and all-sky (Fig. 7f) conditions, especially in regions where the surface albedo decreased (compare Fig. 7b~~,~~ ~~f). Therefore, both changes in surface albedo and in AOT increase the net cooling effect of aerosol particles under clear-sky conditions and decrease the net warming effect under all-sky conditions in the Arctic region. When we compare our results with those by Struthers et al. (2011, averaged over 70-90° N, JJA), we find that the absolute changes in aerosol-radiation interactions are similar despite the different absolute values: while Struthers et al. (2011) report a decrease in the direct aerosol radiative forcing from -0.085 (2000) to $-0.252\,\mathrm{W m^{-2}}$ (2100), we find a decrease from 0.476 (2004) to $0.287\,\mathrm{W m^{-2}}$ (2050). Our values refer to all aerosol particles, those of Struthers et al. (2011) on the other hand only to natural aerosol particles (defined by them as~~). We cannot distinguish between the RF_{ari} induced by surface albedo changes and that induced by changes in aerosols, but we expect that the increase in natural aerosol emissions decreases RF_{ari} since sea salt and ~~mineral dust). This at least partially explains the discrepancy in absolute values since strongly absorbing BC particles are only included in our estimates.~~ sulphate particles are nearly pure scatterers.

The radiative forcing due to BC deposition on snow decreases significantly (see Supplementary Fig. 11) because less snow-covered sea ice and less snow on land exist. However, the radiative forcing due to deposited BC as well as its absolute changes are small compared to other radiative forcings ~~(significant decreases are always below $0.05\,\mathrm{W m^{-2}}$)~~and CREs. This is also displayed in Tables ~~?? and ??~~6 and 7, which show the area-averaged absolute differences in ~~radiative forcings~~ radiation, radiative forcings, and radiative effects north of 60° N and north of 75° N, respectively.

3.1.4 Cloud radiative effects

Not only the aerosol radiative forcing but also CREs change significantly. Using the radiative kernel (RK) method, we first assess how CREs change only as a function of cloud properties (i.e. independent of changes in ~~e.g.~~ surface albedo or surface temperature). In this case, ~~we find no significant changes~~ both the SW and the LW CRE (RK) become stronger in late summer (Tables 6, 7), for example by $-2.2\,\mathrm{W m^{-2}}$ for SW and $+0.88\,\mathrm{W m^{-2}}$ for LW when averaged between 75° and 90° N. In early autumn, changes in ~~SW CRE are not~~ CREs (RK) are significant when averaged over latitudes between ~~60°/~~75° and 90° N (~~see Tables ??, ??). However, the SW CRE shows locally significant changes, e.g. decreases over the central Arctic Ocean or increases along the west coast of Greenland (~~but not over the whole Arctic; see Tables 6, 7), where the SW and LW CREs

(RK) change by $-0.36\,\mathrm{W\,m^{-2}}$ and $-0.96\,\mathrm{W\,m^{-2}}$, respectively. These decreases in the SW CRE (RK) north of 75° N in early autumn (see also Fig. 8c) ~~, which match the changes in cloud cover in Fig. 6~~can be attributed to increases in the cloud optical thickness and low cloud cover (cloud top altitudes below 680 hPa; not shown). In contrast, the negative changes in LW CRE ~~are not significant, neither for individual gridboxes nor when spatially averaged (~~(RK) north of 75° N (see also Fig. 8f~~, Tables ??, ??~~) are due to decreases in the free-tropospheric cloud cover (cloud top altitudes above 680 hPa; not shown).

If we use the standard method for calculating CREs, which considers also impacts due to changes in surface albedo and surface temperature, changes in both SW and LW CRE are much more pronounced over the central Arctic Ocean in early autumn than with the radiative kernel method (Fig. 8b, e). Similarly to ~~aerosol particles~~$\mathrm{RF_{ari}}$, the large changes in SW CRE are mainly caused by the smaller surface albedo (i.e. larger changes in Fig. 8b than in Fig. 8c). In contrast, increases in LW CRE ~~primarly~~ primarily result from increases in surface temperature ~~. The~~ (Supplementary Fig. 6). The significant decrease of LW CRE over the Bering Sea (which only occurs in Fig. 8e and not in Fig. 8f) can also be explained by changes in surface temperature (a decrease in this case). ~~Due to the ice-albedo-feedback, decreases~~ Decreases in surface albedo are highly correlated with increases in surface temperature over the Arctic Ocean because the surface temperature of ice (which can be much lower than 270 K in early autumn, e.g. due to the ice-albedo feedback) changes to the temperature of sea water (minimum temperature of 271.38 K). Furthermore, changes in cloud cover and thickness affect both SW and LW CRE. Changes in SW and LW CRE thus mostly occur at the same locations. Since they are of opposite sign and on the same order of magnitude, they cancel to a large degree ~~, and~~ (Tables 6, 7). While regionally significant decreases and increases occur in the net CRE in early autumn, it shows no significant changes ~~.~~ when averaged between 60°/75° and 90° N.

In late summer, the net CRE decreases significantly from 2004 to 2050 (by $-10\,\mathrm{W\,m^{-2}}$, averaged between 75° and 90° N), i.e. the cooling effect of clouds increases, even though changes in surface albedo are smaller than in early autumn (-0.12 compared to -0.21; averaged between 75° and 90° N). This is because i) the SW component dominates in these months due to the higher zonal zenith angle and ii) the surface temperature over the central Arctic Ocean does not show pronounced increases like in early autumn (~~more sea ice available~~Table 2), therefore not enhancing the LW CRE. The surface temperature even decreases in some regions because melt ponds on ice can have temperatures higher than 271.38 K (but below 273.16 K) in late summer, while the SST is 271.38 K in gridboxes with $0 < \mathrm{SIC} < 1$ (equilibrium conditions, i.e. heat changes lead to changes in SIC, not SST).

Compared with the results by Struthers et al. (2011), our changes in the SW CRE are rather small: averaged between 70° and 90° N (JJA), the radiative effect increases from -63.7 to $-107.7\,\mathrm{Wm^{-2}}$ (i.e. change by $-44\,\mathrm{Wm^{-2}}$) and from ~~-48.0 to $-55.6\,\mathrm{Wm^{-2}}$~~ -47.1 to $-55.4\,\mathrm{Wm^{-2}}$ (i.e. change by ~~$-7.6\,\mathrm{Wm^{-2}}$~~ $-8.3\,\mathrm{Wm^{-2}}$) in their and our simulations, respectively. The larger relative change reported by Struthers et al. (2011) is likely caused by the larger decrease in SIC (and, thus, albedo): while still considerable parts of the Arctic Ocean are covered by sea ice in our simulations in 2050 (especially in June and July), only little sea ice is left in the simuations by Struthers et al. (2011) in 2100.

~~To investigate which absolute estimate of SW CRE might be more appropriate, we compared the simulated~~ For present-day~~CREs from Struthers et al. (2011) and our study with the ones derived by Intrieri et al. (2002) and Shupe and Intrieri (2003) , which are based on the measurements from the Surface Heat Budget of the Arctic (SHEBA) campaign. The SHEBA campaign~~

took place in the Beaufort and Chukchi Seas from October 1997 to October 1998. Intrieri et al. (2002) and Shupe and Intrieri (2003) reported values for CREs at the surface, but since the SW CREs at the TOA and at the surface are fairly similar (Laszlo and Pinker, 1993; Ma , we expect a resembling absolute bias at the TOA. From June to August, our simulated values in the region of the SHEBA campaign are for each month $\approx -20\,\mathrm{Wm^{-2}}$ more negative than the results by Intrieri et al. (2002) and Shupe and Intrieri (2003) . This indicates that our SW CRE might be overestimated, whereas the LW CRE is in good agreement. Provided that the results from the SHEBA campaign are also representative for other Arctic regions and other years than 1998, this suggests that the SW CRE by Struthers et al. (2011) would be even more overestimated than in our simulations. , the absolute estimates of SW CRE by the two models are similarily close to the satellite-derived value by the Clouds and the Earth's Radiant Energy System (CERES), which is $-56.8\,\mathrm{Wm^{-2}}$ averaged over the same months and latitudes for the period July 2005 to June 2015 (Loeb et al., 2018).

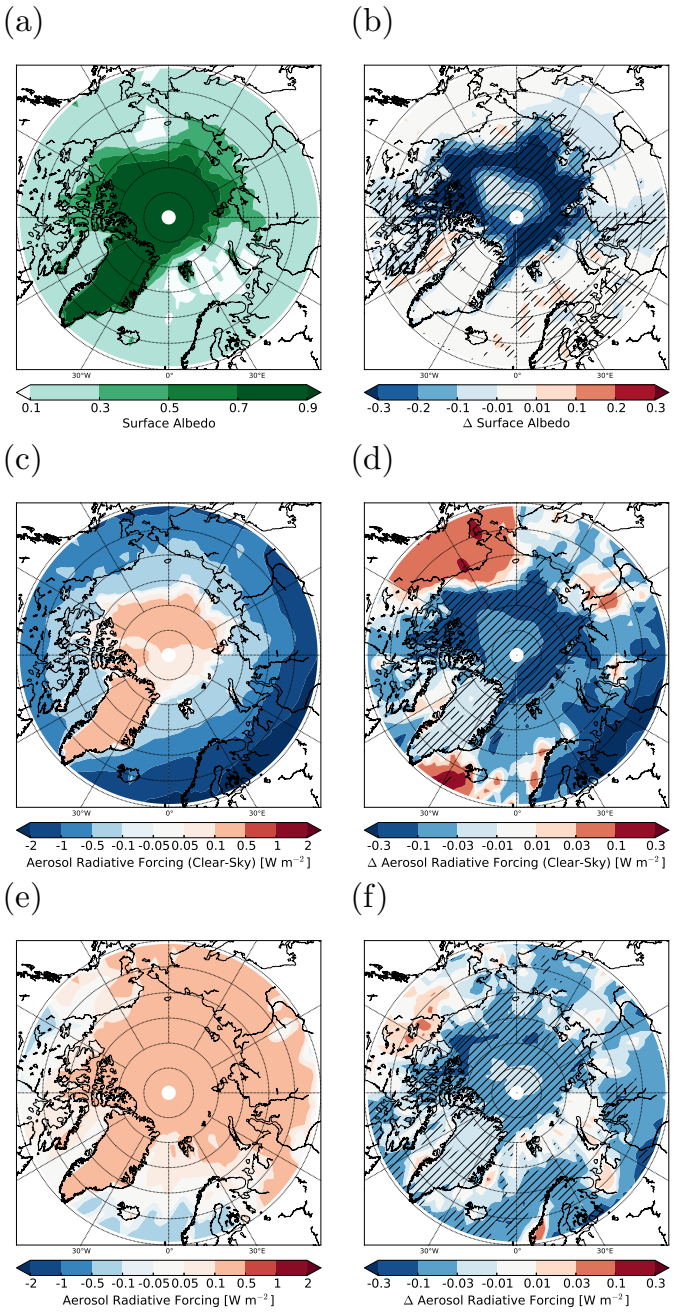

Figure 7. Surface albedo, aerosol net radiative forcing (clear-sky), and aerosol net radiative forcing (all-sky) in 2004 in (a)/(c)/(e) and differences between 2050 and 2004 (i.e. between simulations **arctic_2050_EM2004** and **arctic_2004**) in (b)/(d)/(f) in early autumn (Sep/Oct). ~~Stippled~~ Hatched areas are significant at the 95% confidence level.

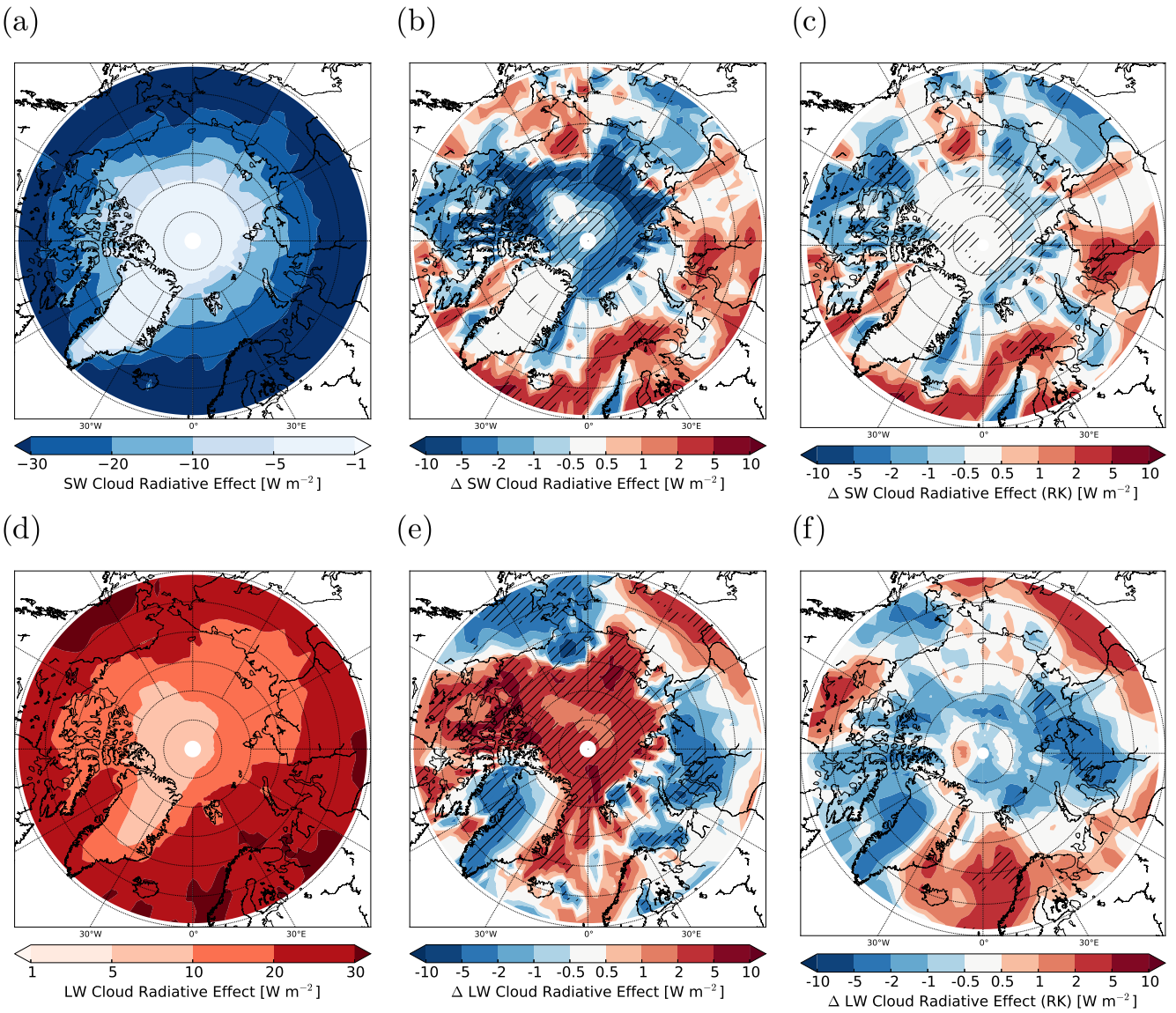

Figure 8. SW and LW CRE in 2004 in (a)/(d) and differences between 2050 and 2004 (i.e. between simulations **arctic_2050_EM2004** and **arctic_2004**) in (b)/(c)/(e)/(f) in early autumn (Sep/Oct). In (b) and (e), the changes in CREs were calculated online from two radiation calls (once with, once without clouds). In (c) and (f), the changes in CREs were calculated with the radiative kernel (RK) method ~~, which does not depend on e.g. changes in surface albedo and surface temperature~~(see text for more details). ~~Stippled~~ Hatched areas are significant at the 95% confidence level.

Table 4. Absolute values for the year 2004 and differences between 2050 and 2004 (i.e. **arctic_2050_EM2004−arctic_2004**) in radiation, radiative forcings, and CREs (in $\mathrm{W\,m^{-2}}$) averaged over all latitudes north of 60° N in late summer (Jul/Aug) and early autumn (Sep/Oct). **arctic_2050_EM2004−arctic_2004** accounts for changes between 2050 and 2004 associated with a warmer climate, which leads to a reduction in SIC and therefore increased natural aerosol emissions. RK stands for radiative kernel method (see text for details). The star (*) denotes changes that are significant at $\alpha = 5\,\%$.

	Late summer (2004)	Late summer (2050 − 2004)	Early autumn (2004)	Early autumn (2050 − 2004)
Net SW radiation	233	3.4*	67	0.70*
Net LW radiation	-231	0.60	-202	-1.7*
RF_{ari}	$12{\cdot}10^{-2}$	$-9.5{\cdot}10^{-2}$*	$9.4{\cdot}10^{-2}$	$-3.4{\cdot}10^{-2}$*
BC deposition	$13{\cdot}10^{-2}$	$0.02{\cdot}10^{-2}$	$1.9{\cdot}10^{-2}$	$0.49{\cdot}10^{-2}$
SW CRE	-67	-4.0*	-26	-0.45*
LW CRE	18	-0.04	21	0.55*
SW CRE (RK)	-67	-2.0*	-26	-0.00
LW CRE (RK)	18	0.92*	21	-0.07

3.2 Impact of additional ship emissions

Future sea ice retreat will enable ships to ~~pass through~~ cross the Arctic Ocean, thus likely leading to enhanced shipping activity in late summer and early autumn. In this section, we will study the influence of these anthropogenic aerosol emissions on aerosol populations, clouds, and their radiative forcings/effects by comparing the simulation **arctic_2050_shipping** with **arctic_2050**.

3.2.1 Aerosol particles

Due to the increase in Arctic ship emissions (tenfold increase of the ship emissions by Peters et al. (2011) in 2050), the burdens of BC and ~~SO_4~~ sulphate are significantly enhanced in late summer (not shown). In early autumn, rises in ship-related aerosol burdens are ~~even~~ more pronounced and also significant for OC ~~. The increases in burden~~ (not shown). The maximum increases in aerosol burdens (see Fig. 9b) occur at the same locations as the ship emissions, but significant increases can spread over a large part of the Arctic (see Fig. 9c), as shown for the example of BC. The largest absolute changes in BC concentration occur near the surface, although significant changes reach altitudes as high as 400 hPa in early autumn (Supplementary Fig. 12d).

Table 5. The same as Table 6 but averaged over all latitudes north of 75° N.

	Late summer (2004)	Late summer (2050 – 2004)	Early autumn (2004)	Early autumn (2050 – 2004)
Net SW radiation	201	12*	29	2.5*
Net LW radiation	-228	0.77*	-196	-4.4*
RF_{ari}	$53 \cdot 10^{-2}$	$-17 \cdot 10^{-2}$*	$15 \cdot 10^{-2}$	$-4.1 \cdot 10^{-2}$*
BC deposition	$21 \cdot 10^{-2}$	$0.32 \cdot 10^{-2}$	$2.0 \cdot 10^{-2}$	$0.99 \cdot 10^{-2}$
SW CRE	-45	-10*	-7.8	-2.2*
LW CRE	9.3	-0.06	13	2.0*
SW CRE (RK)	-45	-2.2*	-7.8	-0.36*
LW CRE (RK)	9.3	0.88*	13	-0.96*

While the changes in natural aerosol emissions (2050 versus 2004) only have a minor influence on the number size distribution (Fig. 3), the impact of increased ship emissions is considerably larger. Figure 10 shows the aerosol number size distributions averaged between ~~70°~~ 75° and 90° N, at both 950 hPa (corresponding to ~~≈ 540 m using the hypsometric equation~~≈ 540 m; Fig. 10a) and 800 hPa (corresponding to ~~≈ 1950 m~~≈ 1950 m; Fig. 10b) for early autumn. At 950 hPa, the number of particles in the nucleation mode ~~decreases, whereas it increases in the accumulation mode~~ largely decreases in both seasons (Fig. 10a). For the Aitken mode, a ~~slight~~ small decrease and a distinct increase occur in late summer (not shown) and early autumn, respectively. The number concentration in the accumulation mode increases to some extent in both late summer and early autumn. At 800 hPa (Fig. 10b), the effect of ship emissions on the aerosol size distribution is smaller than at 950 hPa~~(Fig. 10b). In addition to slight changes in the other three size modes, the number of coarse mode particles increases. However, for the coarse mode, the standard deviation is much larger at than near the surface, which makes this increase very uncertain.~~.

~~We hypothesise that the~~ The additional aerosol particles emitted by ships provide additional surfaces for the condensation of gaseous sulphuric acid~~, which suppresses nucleation and thus decreases the number concentration of~~ . Thus, the vertically integrated condensation rate of sulphate increases where the ship emissions occur (not significant; Supplementary Fig. 13b). The vertically integrated nucleation rate of sulphate shows neither a clear decrease nor a clear increase along the shipping paths (Supplementary Fig. 13d); if the increase in condensation suppressed nucleation (as Fig. 10a suggests), we would expect a decrease in the nucleation rate. However, the vertical cross section of aerosol particles in the nucleation mode ~~.~~ shows that the number concentration indeed decreases significantly near the surface (Supplementary Fig. 13f).

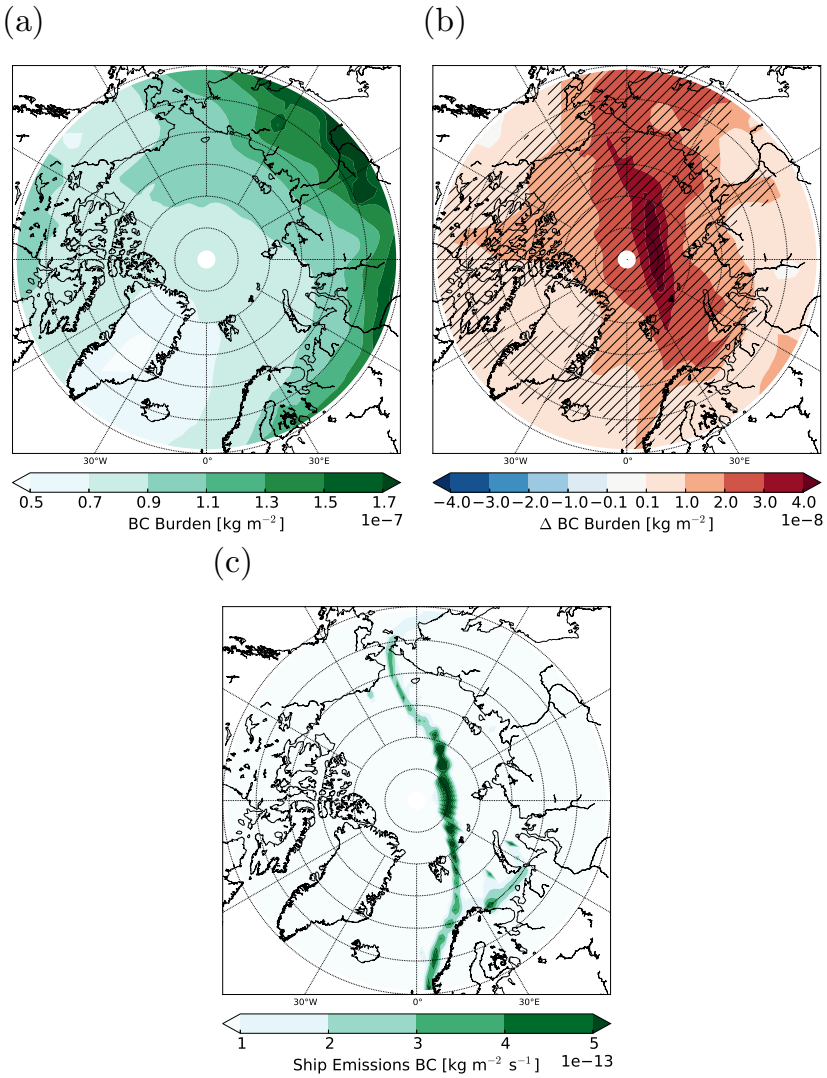

Figure 9. Panel (a) shows the BC burden in 2050 without considering enhanced Arctic ship emissions. Panel (b) shows the difference between a simulation with additional Arctic ship emissions and a simulation without these emissions in 2050 (difference between **arctic_2050_shipping** and **arctic_2050**). ~~Stippled~~ Hatched areas are significant at the 95% confidence level. Panel (c) shows tenfold higher (transit and petroleum-related) ship emissions of BC in 2050 based on the emission inventory by Peters et al. (2011). All values are for early autumn (Sep/Oct).

The number concentrations in the ~~other three size modes~~ accumulation mode (and the Aitken mode in early autumn) increase both by direct emissions and by shifting aerosol particles to larger sizes due to coagulation and condensation. Since ship emissions occur near the surface, the influence at 800 hPa is much smaller than at 950 hPa.

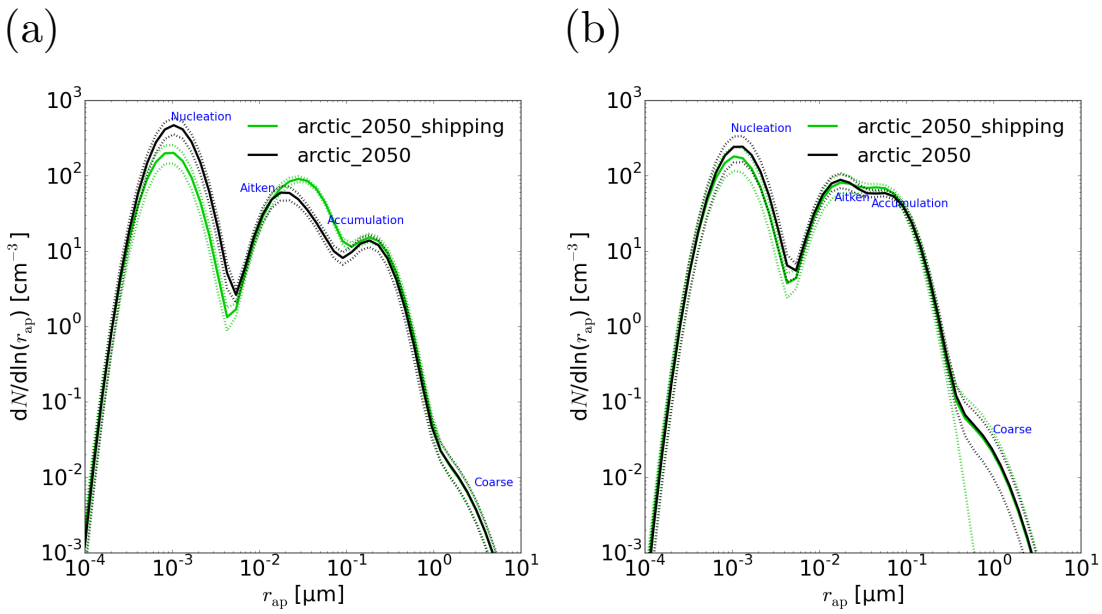

Figure 10. The impact of additional future ship emissions (**arctic_2050_shipping** versus **arctic_2050**) on aerosol number size distributions; N stands for the number concentration (assuming that 1 $kg_{air} \approx 1\ m^3$), r_{ap} for the radius of the aerosol particles. The size distributions are shown for early autumn (Sep/Oct) at 950 hPa (a) and 800 hPa (b), averaged between ~~70°~~ 75° and 90° N. The solid lines denote ensemble means, the dotted lines the subtracted/added standard deviations. Different colors (black, green) stand for different simulations (see legend).

3.2.2 Clouds

Although ship emissions have a larger effect on aerosol burdens and size distributions in early autumn than in late summer, significant aerosol-induced changes in clouds ~~only~~ predominantely occur in late summer. In the following, we will therefore only discuss results for late summer. The ~~impact is mainly restricted to liquid clouds near the surface over the Arctic Ocean. Consistent with the Twomey effect, the CDNC increases~~ CDNC increases (Fig. 11b; increase in CDNC burden by 33% averaged between 75° and 90° N) and the effective radius decreases with additional ship emissions (~~see~~ Fig. 11~~)~~d), consistent with the RF_{aci}. Overall, the increase in CDNC ~~contributes more to the cloud water mass than~~ dominates over the decrease in cloud droplet radius, leading to ~~a somewhat enhanced liquid water content.~~an enhanced LWC (Fig. 11f). We attribute this increase in LWC to a slower collision-coalescence process (cloud adjustments).

Using satellite data, Christensen et al. (2014) studied the effect of ship tracks on both mixed-phase and liquid clouds. In ~~late summer~~ the late summer of 2050, the clouds that are impacted by ships in our simulations are mostly liquid. Therefore, we restrict our comparison to the influence of ships on liquid clouds. Consistent with ~~our results, Christensen et al. (2014)~~ the observations by Christensen et al. (2014), we also found decreases in the effective radius and increases in cloud optical

Table 6. As Table 6 but for **arctic_2050** (absolute values) and **arctic_2050_shipping−arctic_2050** (differences) averaged over all latitudes north of 60° N in late summer (Jul/Aug) and early autumn (Sep/Oct). **arctic_2050_shipping−arctic_2050** considers the impact of an increase in future Arctic ship emissions in 2050.

	Late summer (2050)	Late summer ($2050_{ship} - 2050$)	Early autumn (2050)	Early autumn ($2050_{ship} - 2050$)
Net SW radiation	238	-3.0*	68	-0.46*
Net LW radiation	-231	-0.01	-204	0.32
RF_{ari}	$11 \cdot 10^{-2}$	$0.79 \cdot 10^{-2}$	$4.1 \cdot 10^{-2}$	$1.1 \cdot 10^{-2}$
BC deposition	$12 \cdot 10^{-2}$	$-0.26 \cdot 10^{-2}$	$2.1 \cdot 10^{-2}$	$0.15 \cdot 10^{-2}$
SW CRE	-69	-2.9*	-26	-0.46*
LW CRE	18	-0.04	21	0.35
SW CRE (RK)	-69	-3.4*	-26	-0.46*
LW CRE (RK)	18	0.20	21	0.26

thickness. ~~The liquid water content~~ The relative changes in effective radius are larger in their observations (-20% at cloud top altitude) than in our simulations (-2% to -4% at altitudes below 500 hPa; averaged between 75° and 90°N), whereas changes in cloud optical thickness compare well (+20% in both studies, averaged between 75° and 90°N). The LWP slightly decreases in their analysis (-1%; in-cloud); in contrast, it ~~slightly~~ increases in our simulations (+17%; all-sky, averaged between 75° and 90°N). While our simulated precipitation shows no clear trend, the results by Christensen et al. (2014) suggest that ship emissions delay precipitation by enhancing cloud lifetime. The different results could be explained by the location of the ship tracks analysed by Christensen et al. (2014): the majority of their samples lie between 45° S and 45° N, and only very few datapoints are from the Arctic. Precipitation formation at high ~~and low~~ latitudes differs considerably from that at low latitudes since e.g. convection is usually much more important at low latitudes.

While liquid clouds are significantly impacted by ships in our simulations, this is not the case for cloud ice, neither in late summer nor in early autumn. Theoretically, ship emissions could influence heterogeneous freezing in ECHAM6-HAM2 by several processes, for example:

- The increase in BC emissions could lead to enhanced immersion freezing by BC.

Table 7. The same as Table 8 (impact of additional Arctic shipping) averaged over all latitudes north of 75° N.

	Late summer (2050)	Late summer ($2050_{ship} - 2050$)	Early autumn (2050)	Early autumn ($2050_{ship} - 2050$)
Net SW radiation	213	-3.9*	32	-0.45*
Net LW radiation	-227	-0.47	-200	-0.75
RF_{ari}	$41 \cdot 10^{-2}$	$1.3 \cdot 10^{-2}$	$11 \cdot 10^{-2}$	$0.52 \cdot 10^{-2}$
BC deposition	$19 \cdot 10^{-2}$	$0.64 \cdot 10^{-2}$	$2.5 \cdot 10^{-2}$	$-0.02 \cdot 10^{-2}$
SW CRE	-57	-3.7*	-9.9	-0.38*
LW CRE	9.1	-0.23	15	0.61
SW CRE (RK)	-57	-4.4*	-9.9	-0.35*
LW CRE (RK)	9.1	0.18	15	0.46

- The increased SO_2 emissions could shift some dust particles from the insoluble to the internally mixed mode, which shifts contact freezing to immersion freezing, i.e. to colder temperatures~~(as found by e.g. Hoose et al., 2008)~~, as found by e.g. Hoose et al. (2008).
- Decreases in the droplet radius would decrease the contact freezing rate.
- Increases in the CDNC would increase the contact freezing rate.

The last two effects might partly cancel each other since a larger number concentration of CCN is expected to simultaneously decrease the droplet radius and increase the CDNC. However, also the first two points seem to be irrelevant as ship emissions have no significant impact on ~~ice clouds~~ cloud ice in our simulations. To better understand why and gain some insights into the importance of the different heterogeneous freezing processes, we calculated the number of ice crystals that freeze ~~in~~ via each of these processes (Fig. 12a, c, e). Immersion freezing by dust is the dominant freezing process in the Arctic in late summer (Fig. 12c). However, contact freezing by dust is more important near the surface since it can induce freezing at higher temperatures than immersion freezing (Fig. 12a). With additional ship emissions, the number of ice crystals formed by contact freezing decreases near the surface and increases at higher altitudes (Fig. 12b). Since the relative changes in CDNC are larger than the relative changes in droplet radius (which would increase the contact freezing rate), we suspect that contact freezing

near the surface is reduced by shifting more dust particles to the internally mixed modes. This is consistent with the slight (non-significant) increase in immersion freezing occurring near the surface (Fig. 12d).

Compared to dust, BC initiates freezing only in very few cloud droplets (Fig. 12e) ~~. Furthermore,~~ because its influence is mainly restricted to high altitudes where temperatures are sufficiently low to initiate freezing. However, BC particles from ships are emitted near the surface. ~~Although~~ Therefore, the largest increases in BC concentrations also occur near the surface (Supplementary Fig. 12b). As a consequence, BC immersion freezing is ~~somewhat~~ slightly enhanced near the surface (Fig. 12f), but absolute changes are orders of magnitudes smaller than the decreases in contact freezing of dust. These findings lead to the conclusions that i) BC immersion freezing is largely not affected because of the low altitude of ship emissions, ii) even if it were, it would hardly matter because dust is by far the dominant INP, and iii) SO_2 emissions from ships lead to a slight shift from contact to immersion freezing near the surface, thus rather leading to a non-significant decrease in cloud ice at low altitudes.

Heterogeneous freezing is still an active field of research, and contradictory evidence exists concerning the ability of combustion aerosols to act as INPs (Kanji et al., 2017). Laboratory results suggest that soot starts initiating freezing at temperatures $\leq -30\,^{\circ}\mathrm{C}$ (Kanji et al., 2017, Fig. 1-7). On the other hand, Thomson et al. (2018) found an increase in INP concentrations in ship tracks at higher temperatures. The increases were small at temperatures around $-20^{\circ}\mathrm{C}$, moderate at $-25^{\circ}\mathrm{C}$ ($\approx +0.5\,\mathrm{L}^{-1}$; saturation ratio of 1.22), and quite pronounced at $-30^{\circ}\mathrm{C}$ ($\approx +2\,\mathrm{L}^{-1}$; saturation ratio of 1.32). The ship plumes were measured near the port of Gothenburg (57.7° N, 11.8° E) in 2013 and 2014, and the meteorology in general represented climate conditions of the late-autumn maritime North. If ship exhaust (not necessarily the BC particles) can indeed induce freezing at higher temperatures than in the laboratory-based BC-parameterisation used in our model, the impact on cloud ice could be larger than in our simulations, especially in early autumn when temperatures are colder.

3.2.3 Aerosol radiative forcings

The higher aerosol burdens due to ship emissions lead to ~~significantly enhanced AOTs . Changes~~ enhanced AOTs (significant increase from $1.4 \cdot 10^{-2}$ to $2.0 \cdot 10^{-2}$ in late summer and insignificant increase from $1.4 \cdot 10^{-2}$ to $1.5 \cdot 10^{-2}$ in early autumn; averaged between 75° and 90°N). Changes induced by additional ship emissions are on the same order of magnitude as the changes caused by additional sea salt and DMS emissions from 2004 to ~~2050.~~ 2050 ($\approx +0.2 \cdot 10^{-2}$). In contrast to the ~~sea salt and DMS induced changes , which hardly affect aerosol absorption~~ changes in aerosol absorption from 2004 to 2050 (no significant changes in late summer; decrease in early autumn), ship emissions ~~increase~~ lead to pronounced and significant increases in the aerosol absorption optical thickness ~~pronouncedly~~(from $1.12 \cdot 10^{-3}$ to $1.19 \cdot 10^{-3}$ in late summer and from $0.83 \cdot 10^{-3}$ to $1.00 \cdot 10^{-3}$ in early autumn; averaged between 75° and 90°N). This is not surprising since OC and predominantely BC are important absorbers of sunlight.

In late summer, the SW component clearly dominates changes in the net ~~aerosol radiative forcing~~$\mathrm{RF_{ari}}$ (e.g. $+13\,\mathrm{mW\,m^{-2}}$ in SW compared to $+0.40\,\mathrm{mW\,m^{-2}}$ in LW under all-sky conditions; averaged between 75° and 90°N). Under clear-sky conditions, the ship emissions induce a pronounced cooling (i.e. $\mathrm{RF_{ari}}$ decreases; see Fig. 13b). This cooling reverses to a non-significant warming under all-sky conditions (Fig. 13d). Again, this shows that the scattering of aerosol particles becomes

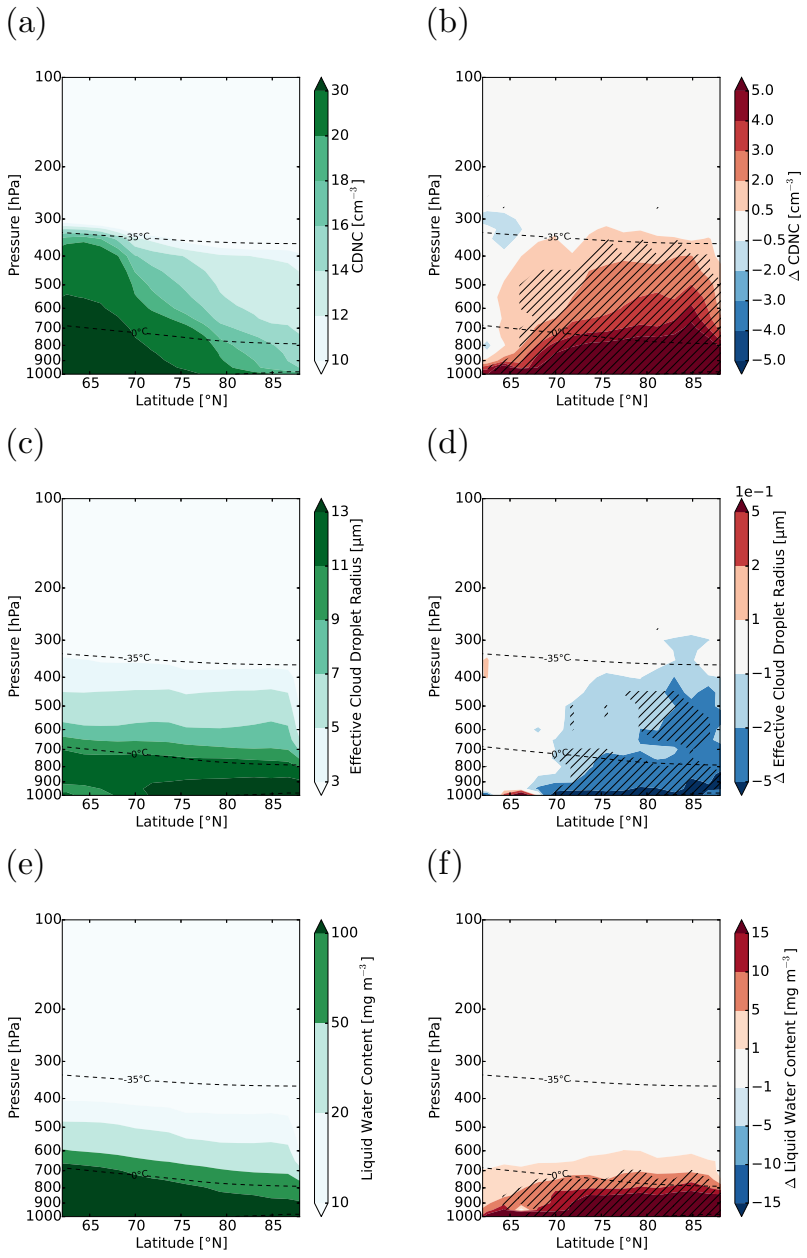

Figure 11. CDNC~~and~~, effective cloud droplet radius, and LWC in late summer (Jul/Aug; in-cloud values): (a)/(c)/(e) show the absolute values for 2050 (reference), (b)/(d)/(f) the difference between a simulation with enhanced ship emissions and the reference simulation (difference between **arctic_2050_shipping** and **arctic_2050**). ~~Stippled~~ Hatched areas are significant at the 95% confidence level. The dashed lines show the $0^{\circ}\mathrm{C}$ ~~–~~and the $-35^{\circ}\mathrm{C}$ ~~isotherms~~isotherms.

less important when the scattering of clouds is considered as well, and that the aerosol absorption can be enhanced in the presence of clouds.

In early autumn, changes in the SW component still dominate ~~the aerosol radiative forcing~~ changes in the RF_{ari} in the region of shipping activity ~~under clear-sky conditions. However, increases in the LW effect can locally be as important~~ (e.g. ~~northwards of 80° N~~ $+8\,mW\,m^{-2}$ in SW compared to $+3 mW\,m^{-2}$ in LW under all-sky conditions; averaged between 75° and 90°N). Under ~~all-sky~~ clear-sky conditions, the ~~SW aerosol radiative forcing does not change significantly, whereas the LW aerosol radiative forcing shows small but significant increases over a large part of the Arctic. However, like in late summer~~ship emissions lead to locally significant decreases in RF_{ari} (see Supplementary Fig. 14b). Under all-sky conditions, changes in ~~the net aerosol radiative forcing~~ net RF_{ari} are not significant ~~except for averaged values north of 75° N (see Table ??~~(Table 9).

In early autumn, the BC deposition on snow leads to a small but significant warming over part of the Arctic Ocean (~~at most $0.01 - 0.05\,W\,m^{-2}$;~~ see Fig. 14~~d~~f). Although these changes are pronounced in relative terms, they are more than one order of magnitude lower in absolute terms compared to the enhanced cooling by clouds, which is discussed in the next section: averaged between 60° and 90° N, ~~deposited BC causes a warming of $0.018\,W\,m^{-2}$ in late summer~~the radiative forcing of deposited BC insignificantly increases by $1.5 \cdot 10^{-3}\,W\,m^{-2}$ in early autumn, while the SW CRE ~~induces a cooling of $0.99\,W\,m^{-2}$ (Table ??~~is significantly enhanced by $-2.9\,W\,m^{-2}$ in late summer (Table 8).

Based on the future Arctic ship emissions by Corbett et al. (2010), Dalsøren et al. (2013) estimated how short-lived atmospheric pollutants might change by 2030. Meteorology, sea ice extent, and emissions not related to ships were not changed between 2004 and 2030 in their simulations. Therefore, we compare our simulated changes which are only due to shipping (change from **arctic_2050** to **arctic_2050_ship**) with their results. In their high emission scenario, BC and OC annual ship emissions increase in the Arctic by 2030 (BC by a factor of ≈ 5 and OC by a factor of ≈ 2), whereas SO_2 emissions slightly decrease (by $\approx 4\%$). In our simulations, annual Arctic BC, OC, and SO_2 ship emissions increase by factors of 11, 10, and 7, respectively. Averaged between 60° and 90° N and over August, September, and October, Dalsøren et al. (2013) find that the radiative forcing of aerosols overall increases: $+5\,mW\,m^{-2}$ for sulphate, $+5$ to $+6\,mW\,m^{-2}$ for BC, and nearly no changes for OC. The sum of these values is larger than the value that we find ($+5.7\,mW\,m^{-2}$ averaged over the same time period and area) although our increases in ship emissions are higher. It is possible that the radiative forcing of all aerosols is more positive in the study by Dalsøren et al. (2013) because of the different SO_2 emissions: in our simulations, the SO_2 emissions increase, which leads to cooling. In contrast, the SO_2 emissions in the study by Dalsøren et al. (2009) slightly decrease, which leads to a small positive forcing. Furthermore, the effect of clouds on RF_{ari} might differ between the simulations by Dalsøren et al. (2009) and our simulations. The changes induced by deposited BC are $\approx 1\,mW\,m^{-2}$ in both the study by Dalsøren et al. (2009) and in our simulations. While the increase in BC emissions is much larger in our simulations, less snow is available in 2050 compared to 2004.

3.2.4 Cloud radiative effects

In late summer, aerosol particles from ships lead to more but smaller cloud droplets ~~, which reflect more~~ and an enhanced LWC (ERF_{aci}), which increases the reflection of solar radiation. Thus, we see an enhanced cooling effect of clouds in most areas

where the CDNC burden increases (Fig. 14b, d), i.e. the SW CRE becomes significantly more negative (~~≈ 2 – 20 W m^{-2}~~≈ $-3.7\,\mathrm{W\,m^{-2}}$, averaged between 75° and 90° N). Changes in the LW CRE are smaller in terms of absolute amount~~and not as~~, not consistently spatially correlated with ship emissions~~. However, changes are not significant , neither in the SW nor in the LW~~, and not significant (not shown). We additionally analysed the different contributions to the changes in CREs from cloud cover, cloud top altitude, and cloud thickness (see Fig. 15~~; the residual in (g) and (h) shows~~). The residuals in Fig. 15g and 15h show what can be attributed to neither cloud cover, nor cloud top altitude, nor cloud thickness~~)~~; it should ideally be zero. While the changes in CRE caused by changes in cloud cover and cloud top altitude ~~is~~ are not significant (Fig. 15a-d), the increase in cloud optical thickness leads to significant decreases and increases in the SW and LW CRE, respectively (Fig. 15e, f). Averaged between 75° and 90° N, the increased optical thickness changes the SW CRE by ~~−2.89 W m^{-2}~~ $-4.6\,\mathrm{W\,m^{-2}}$ and LW CRE by ~~0.27 W m^{-2}~~ $0.52\,\mathrm{W\,m^{-2}}$ in late summer (significant). When we partition the contributions from low and free-tropospheric clouds (defined as clouds with a cloud top altitude below/above the altitude of 680 hPa), we find that ~~significant changes induced by~~ 74% of the changes in SW cloud optical thickness ~~only~~ occur in low clouds. This is not surprising considering that the ship emissions occur near the surface. ~~The increases in low cloud optical thickness change the SW and LW CRE by −2.10 W m^{-2} and 0.08 W m^{-2}, respectively~~

Possner et al. (2016) studied the influence of model resolution on ship-induced aerosol-cloud interactions and CREs (marine stratocumuli). They found that the changes in SW CRE were overestimated by a factor of 2.6 with the coarser model resolution ($\Delta x = 50$ km, $\Delta t = 180$ s) compared with the higher model resolution ($\Delta x = 1$ km, $\Delta t = 20$ s). In case this finding is generally applicable to numerical models, it could imply that the SW CRE is also overestimated in our simulations.

In the study by Dalsøren et al. (2013), aerosol-cloud interactions lead to much smaller changes in radiative forcing ($-2\,\mathrm{mW\,m^{-2}}$; averaged between 60° and 90° N and over August, September, and October) than in our simulations ($-0.85\,\mathrm{W\,m^{-2}}$; averaged over the same period and space). This is expected because our changes in future Arctic aerosol ship emissions are considerably larger than in Dalsøren et al. (2009). Furthermore, it should be noted that Dalsøren et al. (2009) calculate $\mathrm{RF_{aci}}$ using an empirical relationship that estimates CDNC from aerosol concentrations. In our case, CCN are calculated based on Köhler theory and we consider fast adjustments, i.e. report $\mathrm{ERF_{aci}}$ instead.

To summarise, ship emissions lead to a ~~significant but weak warming~~ locally significant but very weak positive radiative forcing over the central Arctic Ocean in early autumn caused by absorption of deposited BC on snow. In contrast, the direct impact of aerosol particles on the net radiation ($\mathrm{RF_{ari}}$) is not significant. The changes in CREs are ~~also not significant but indicate~~ significant and show that aerosol particles ~~seem to~~ enhance the cooling effect of clouds in late summer. ~~This is confirmed when we partition the CREs from different components:~~ When we partition CRE into its different components, we find no significant radiative changes induced by changing cloud top altitude or cloud cover, but the cloud optical thickness increases and ~~leads to a~~ is responsible for the significant net cooling. ~~Due to the large variability in clouds (e.g. cloud cover), we do not find a significant signal when looking at all effects together.~~ Since the cooling induced by aerosol-cloud interactions exceeds the warming of deposited BC by at least one order of magnitude, ship emissions ~~can be expected to~~ of aerosols and their precursor gases overall induce a ~~local cooling~~ cooling in our simulations.

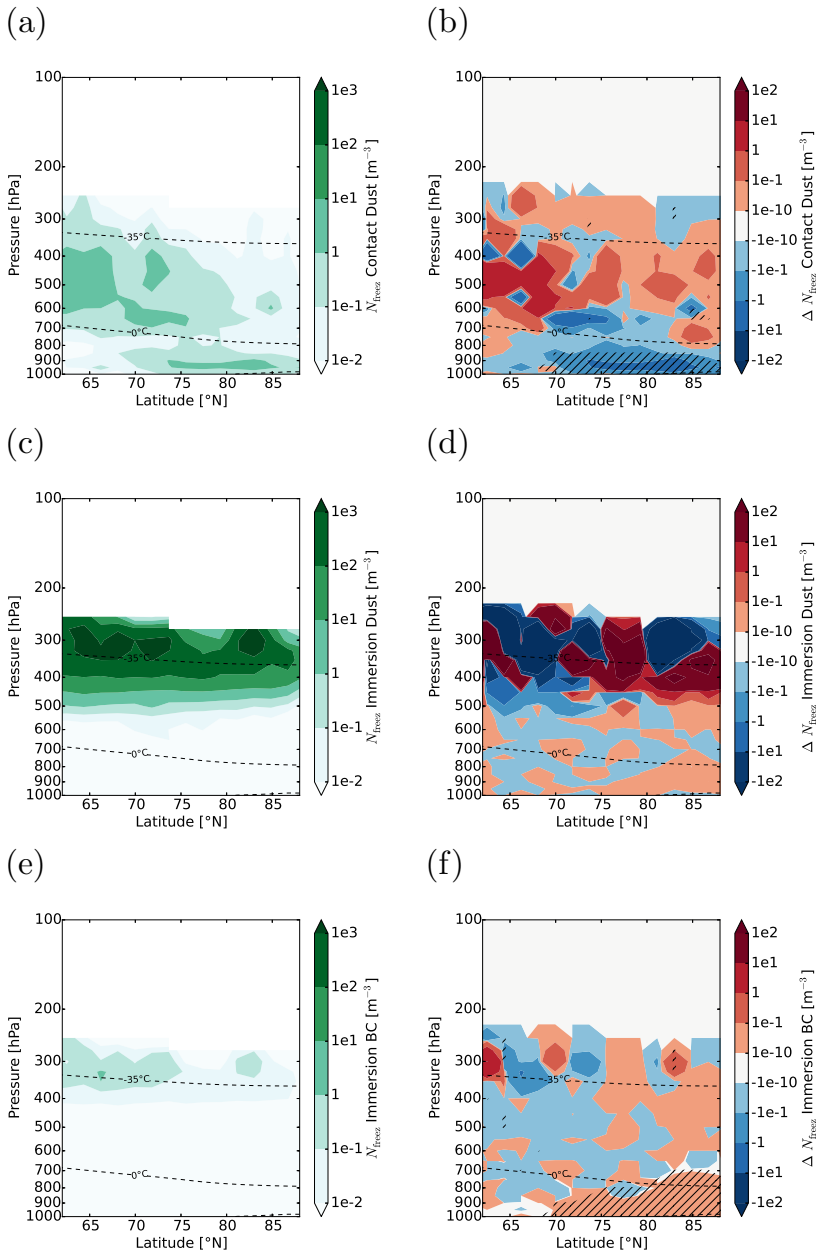

Figure 12. Number of cloud droplets ~~frozen~~ that freeze heterogeneously per timestep (N_{freez}) in 2050: (a)/(b) contact freezing by dust, (c)/(d) immersion freezing by dust, (e)/(f) immersion freezing by black carbon in late summer (Jul/Aug). On the left side, absolute values for 2050 (reference) are shown. On the right side, the difference between a simulation with enhanced ship emissions and the reference simulation is displayed (difference between **arctic_2050_shipping** and **arctic_2050**). Note that the ~~colorbar~~ scale is logarithmic and that the lowest bin had to be decreased to 10^{-10} to display statistically significant increases in immersion freezing by BC. ~~Stippled~~ Hatched areas are significant at the 95% confidence level. The dashed lines show the $0^{\circ}\mathrm{C}$ ~~-~~and the $-35^{\circ}\mathrm{C}$ ~~-isotherms. Note that the shown~~ isotherms~~are a zonal and temporal average, hence ice in mixed-phase clouds can exist at altitudes below the $0^{\circ}\mathrm{C}$-isotherm and above the $-35^{\circ}\mathrm{C}$-isotherm~~.

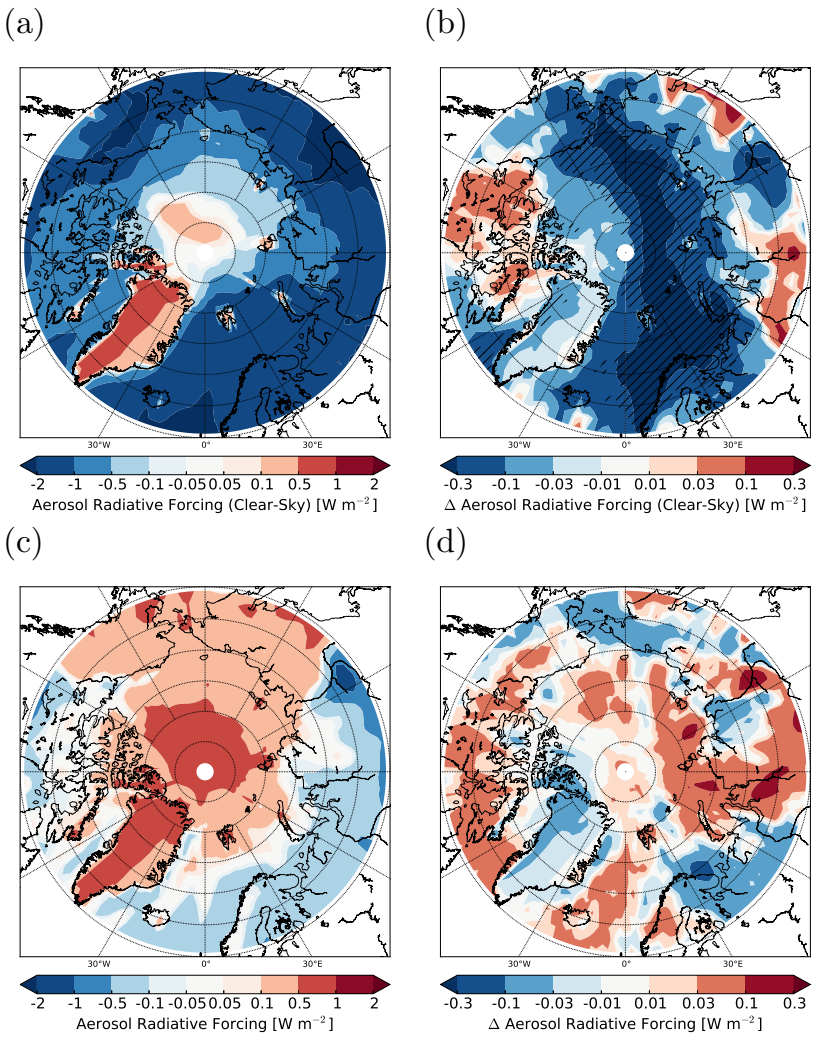

Figure 13. Aerosol radiative forcing in late summer (Jul/Aug) 2050: (a)/(b) under clear-sky and (c)/(d) under all-sky conditions. On the left side, absolute values for 2050 (reference) are shown. On the right side, the difference between a simulation with enhanced ship emissions and the reference simulation is displayed (difference between **arctic_2050_shipping** and **arctic_2050**). ~~Stippled~~ Hatched areas are significant at the 95% confidence level.

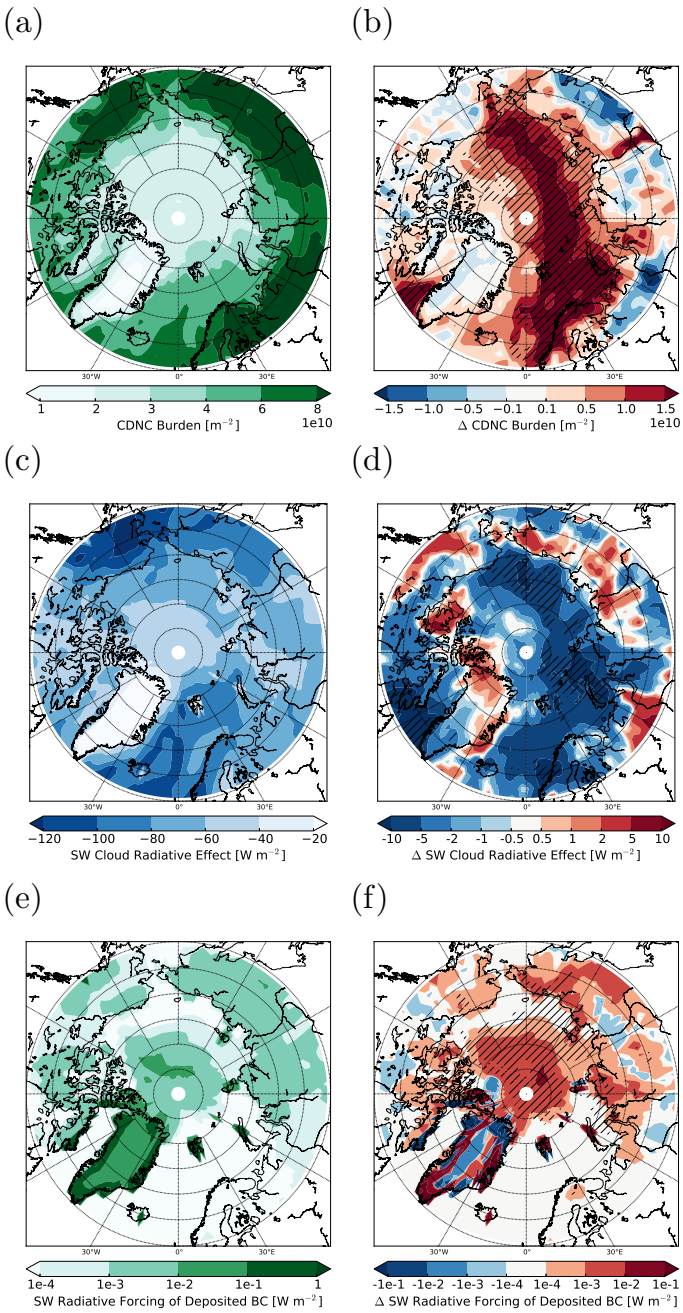

Figure 14. The impact of additional ship emissions in the Arctic on: (b) in-cloud CDNC burden, (d) SW CRE, and (f) radiative ~~effect of~~ forcing due to BC deposition on snow. In (a), (c), and (e), the reference without additional ship emissions is shown (**arctic_2050**). ~~Stippled~~ Hatched areas are significant at the 95% confidence level. (a) to (d) are shown for late summer (Jul/Aug), (e) and (f) for early autumn (Sep/Oct~~; seasons where pronounced changes occur~~). Note that the scale in e) and f) is logarithmic.

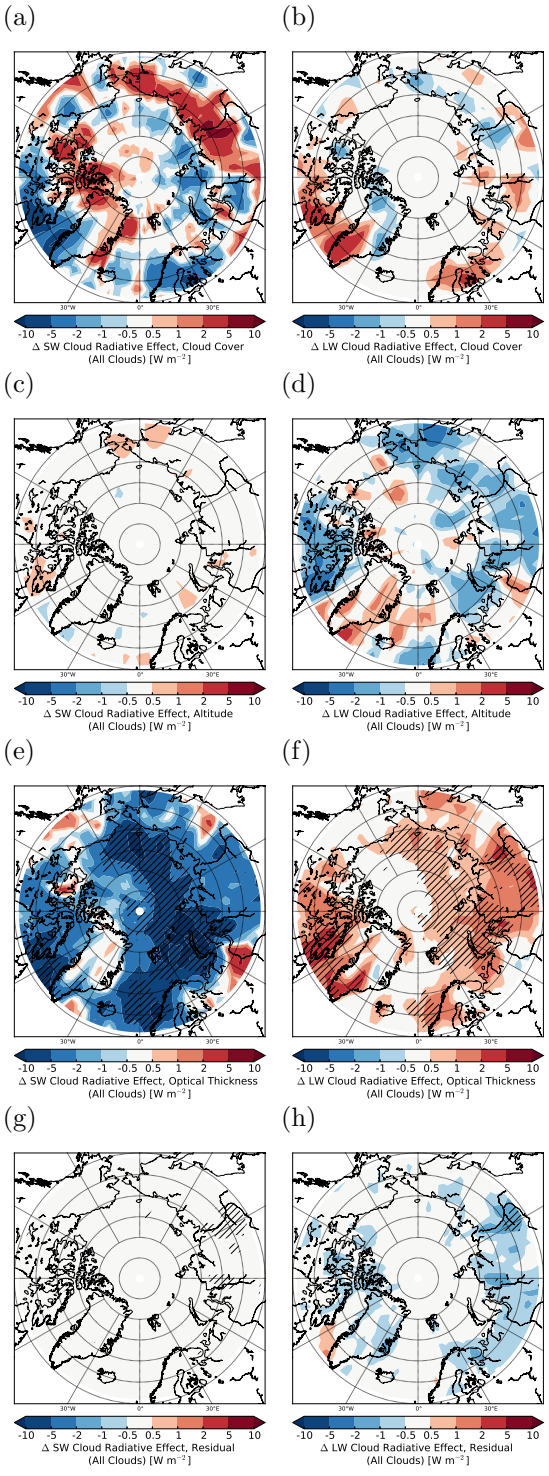

Figure 15. Different contributions to the changes in SW (left) and LW (right) CREs in late summer (Jul/Aug) caused by enhanced shipping: contribution from changes in (a)/(b) cloud cover, (c)/(d) cloud top altitude, and (e)/(f) cloud optical thickness. In (g)/(h), the residual is shown~~, which cannot be attributed to one of the three mentioned contributions (would ideally be zero)~~. ~~Stippled~~ Hatched areas are significant at the 95% confidence level.

4 Summary and conclusions

The main goal of this work was to analyse aerosol-cloud, aerosol-radiation, and cloud-radiation interactions in a warming Arctic when sea ice extent diminishes in late summer and early autumn. Simulations with ECHAM6-HAM2 were conducted for the years 2004 and 2050. We also estimated the impact of enhanced future Arctic shipping activity on climate.

Our results suggest that the future decrease in summer Arctic SIC will significantly increase ~~aerosol~~ sea salt and DMS burdens in the Arctic due to enhanced emissions~~of sea salt as well as DMS. The increase in CCN~~. Both changes in aerosols and meteorology will lead to enhanced CDNCs. Furthermore, not only the number concentration but also the size of cloud droplets will generally increase because of higher specific humidities leading to thicker clouds. In late summer, the net CRE at the TOA will become more negative mainly because of the decrease in surface albedo associated with melting of sea ice. Also ~~the aerosol radiative forcing will decrease~~ RF_{ari} will decrease in late summer and early autumn mainly as a consequence of sea ice melting~~and enhanced aerosol optical thickness in late summer and early autumn~~. The decrease in both net CRE and ~~aerosol radiative forcing (negative temperature feedbacks)~~ RF_{ari} might delay Arctic warming to some extent.

The simulated LWP, cloud cover, CREs, and surface concentrations of BC and sulphate under present-day conditions compare well with Arctic observations. However, our model has a low bias in AOT and cloud ice, which could impact the simulated absolute changes in the radiative forcings and the CREs. Furthermore, intermodel differences in sea salt emissions are large (de Leeuw et al., 2011), and so are the differences between our results and other modelling studies that investigated changes in natural aerosols with declining sea ice. This highlights that the results from this study – as from any climate model study projecting the future – are uncertain.

Arctic ship emissions related to transport and oil/gas extraction have a negligible impact on clouds and radiation in our simulations. Only when we increase the ship emissions of Peters et al. (2011) by a factor of ten is the signal-to-noise ratio sufficiently large to detect ship-induced changes. In this case, the AOT significantly increases ~~on~~ by the same order of magnitude as natural AOT changes from 2004 to 2050. ~~The net aerosol radiative forcing~~ RF_{ari} shows only minor, insignificant changes in the presence of clouds, though. An increase in BC deposition on snow leads to a very small local warming in early autumn. Meanwhile, ~~a Twomey effect~~ ERF_{aci} induces a cooling in late summer. ~~Although this Twomey effect is statistically only significant for radiative effects associated with changes in cloud optical thickness, the~~ The magnitude of changes in ~~radiation~~ ERF_{aci} are considerably larger than those induced by the deposition of BC on snow, implying that ship emissions might overall induce a cooling. In our simulations, only liquid clouds show significant changes with increased ship emissions, while cloud ice is unaffected. Considering the large uncertainty of heterogeneous freezing processes, this result needs to be regarded with caution.

Compared to other changes (such as the decrease in surface albedo or the increase in natural aerosol emissions), ship emissions of aerosols and their precursor gases seem to have a small effect on climate considering that we scaled the emissions up by one order of magnitude. ~~Such high Arctic ship emissions are more likely to occur in years later than 2050 when large population and economic demand could lead to further increases in transit and petroleum-related shipping.~~ However, even though this study suggests that Arctic ship emissions of aerosols and their precursor gases might have a negli-

gible or slightly beneficial impact on climate, they will also increase air pollution and might disturb local flora and fauna. Furthermore, this study does not account for ship-induced changes in greenhouse gases (e.g. O_3), which are also important forcers (Dalsøren et al., 2013; AMAP Assessment, 2015). More studies are required to confirm or ~~object the results found in~~ refute the findings of this work as well as to explore further ship-related environmental impacts.

5 Code availability

The code is available upon request.

6 Data availability

The data is available upon request.

Appendix A: Map of Arctic Seas

As a help for readers not familiar with the Arctic Ocean, Fig. A1 shows its most important regional seas. Furthermore, some land masses are labelled for better orientation.

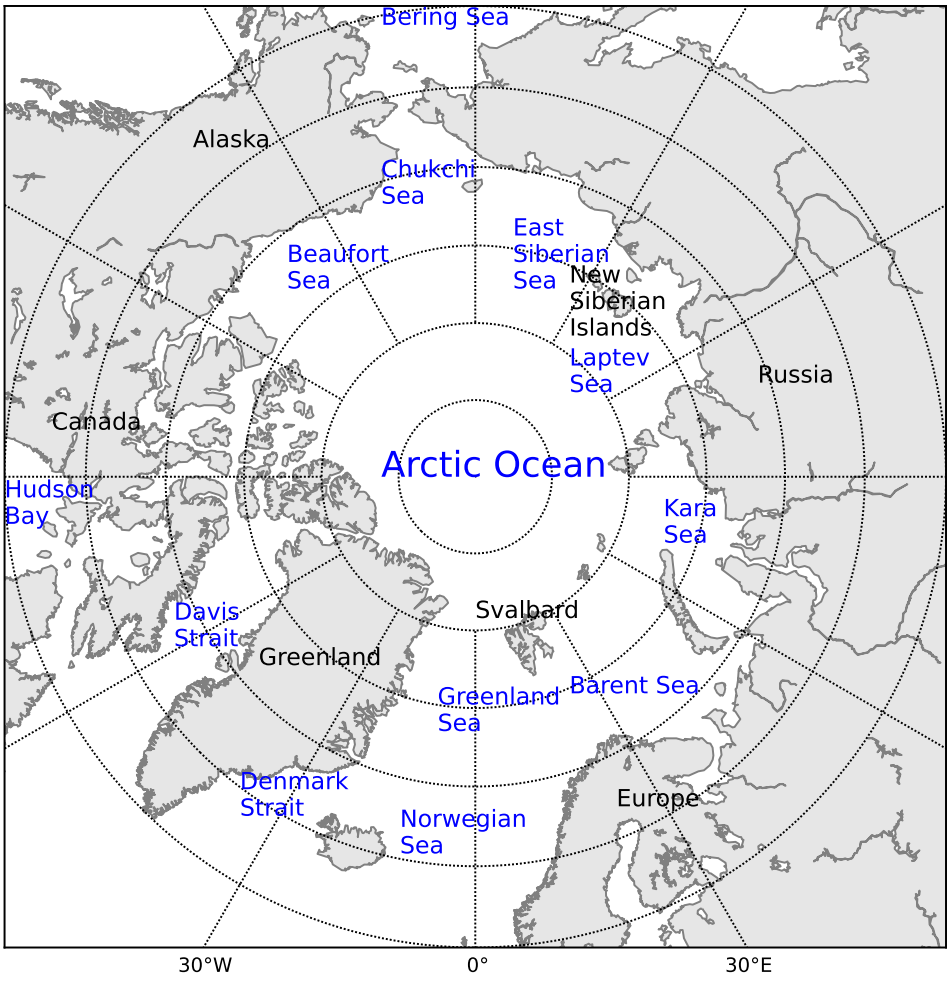

Figure A1. The Arctic Ocean and regional seas are labelled in blue, land masses in black.

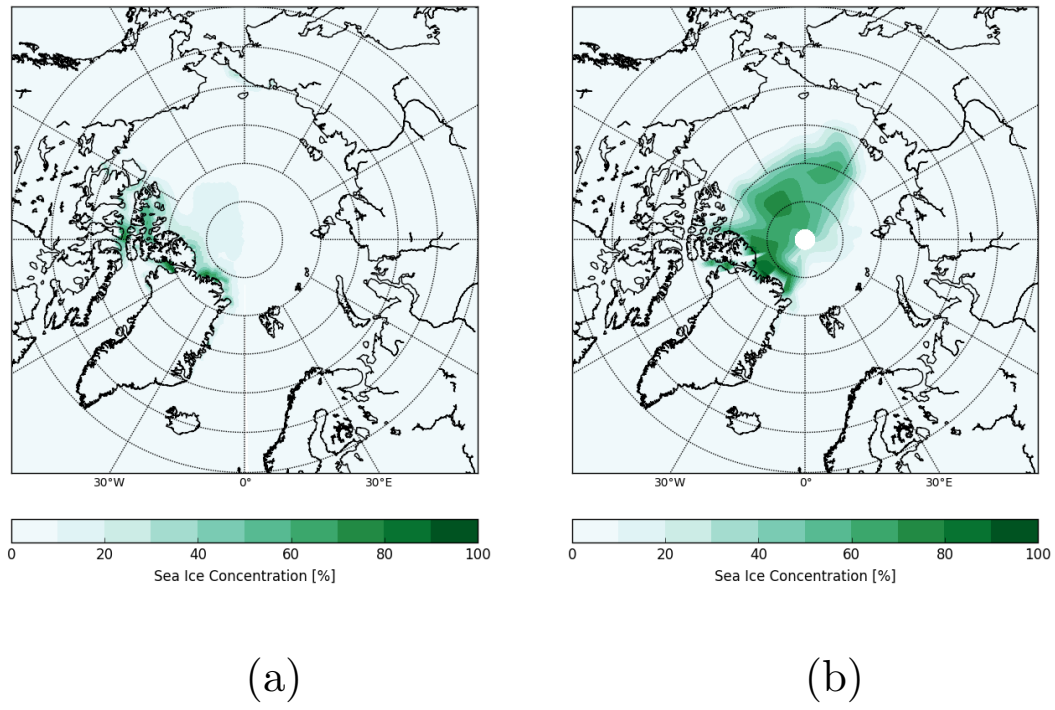

Figure A2. SIC in 2050 for (a) NCAR-CCSM3 in September (average over 5 ensemble members) and (b) MPI-ESM in October (the ensemble member used in this study).

Appendix B: Comparison of sea ice between MPI-ESM and NCAR-CCSM3

Here we compare the sea ice used as input for the study of Peters et al. (2011) with our prescribed sea ice from MPI-ESM. With that we want to ensure that the ship emissions – which explicitly depend on the sea ice thickness and concentration – are compatible with the sea ice used in our study. Peters et al. (2011) used a 5-year running average of the NCAR-CCSM3 model to calculate future sea ice conditions (scenario A2). Instead of averaging over years, we calculated the mean over the 5 ensemble members of NCAR-CCSM3 from CMIP3 for our comparison, which should give similar results. For their calculations, Peters et al. (2011) chose the months March, June, September, and December to represent each season. In our model, we prescribe the sea ice monthly because this is more realistic. Therefore, we will compare the sea ice in July from MPI-ESM with the sea ice in June from NCAR-CCSM3 (used in the calculation for ship emissions in July) and the sea ice from August to October from MPI-ESM with the sea ice in September from NCAR-CCSM3. For this comparison, we will focus on the regions where most Arctic ship emissions are projected to occur in the future.

The sea ice thickness is generally thinner in MPI-ESM than in NCAR-CCSM3. The opposite is the case for the sea ice extent, which is larger in MPI-ESM than in NCAR-CCSM3. In August and October, the SIC in MPI-ESM is higher than the

NCAR-CCSM3 September value (used by Peters et al. (2011) for August, September, and October). At the locations of the ship tracks, differences are most pronounced north of the New Siberian Islands, where the SIC reaches up to 60-70% in MPI-ESM, whereas basically no sea ice is left in NCAR-CCSM3 (see Fig. A2). However, with an extended use of ice breakers, ships can pass despite the higher SIC. Furthermore, the area where this larger SIC for MPI-ESM occurs is rather small, as the SIC in MPI-ESM rapidly decreases towards the New Siberian Islands and the Russian coast. By slightly changing the shipping routes, most of the additional expenses linked to SIC (i.e. to ice breakers, which are included in the cost-benefit analysis of Peters et al., 2011) would be saved. We therefore expect that costs associated with breaking and/or bypassing sea ice are small and should not considerably change the ship emissions derived by Peters et al. (2011).

Appendix C: Significance test for cloud feedback

The cloud feedback is calculated using radiative kernels. These kernels are calculated as *differences* of two simulations, here represented by the vectors $\boldsymbol{a} = \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ \dots \\ a_n \end{pmatrix}$ and $\boldsymbol{b} = \begin{pmatrix} b_1 \\ b_2 \\ b_3 \\ \dots \\ b_n \end{pmatrix}$, where n is the number of samples. In our case, we could not simply use a one sample t-test upon the differences $\boldsymbol{a} - \boldsymbol{b}$ because the differences are calculated from ~~10~~ 20 *independent* samples (i.e. years) with different standard deviations for the different simulations. Instead, we reconstructed from the following differences the standard deviation of $\boldsymbol{a}$, the standard deviation of $\boldsymbol{b}$, and the difference between the means of $\boldsymbol{b}$ and $\boldsymbol{a}$:

- For standard deviation of $\boldsymbol{a}$: calculate standard devation of $\begin{pmatrix} b_1 - a_1 \\ b_1 - a_2 \\ \dots \\ b_1 - a_n \end{pmatrix}$.
- For standard deviation of $\boldsymbol{b}$: calculate standard devation of $\begin{pmatrix} a_1 - b_1 \\ a_1 - b_2 \\ \dots \\ a_1 - b_n \end{pmatrix}$.
- For difference between the means of $\boldsymbol{b}$ and $\boldsymbol{a}$: $\frac{b_1+b_2+\dots+b_n}{n} - \frac{(a_1+a_2+\dots+a_n)}{n} = \frac{b_1-a_1+b_2-a_2+\dots+b_n-a_n}{n}$, i.e. we calculated the kernels between b_1 and a_1, ..., b_n and a_n and calculated the average of these differences.

With this information, we could calculate the p-values using the Welch's test for each gridpoint and control the FDR as described in Sect. 2.4.

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

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