### **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, 1.23 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<sup>-3</sup> 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<sup>-3</sup> 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 Biag, 2005; Chang et al., 2011). However, since the Arctic is a remote environment with low aerosol concentrations, observations show that the value 40 cm<sup>-3</sup> is often undershot in this region: between July 15<sup>th</sup> and September 23<sup>th</sup>, Bigg and Leck (2001) measured daily median CCN concentrations between 15 and 50 cm<sup>-3</sup> at a supersaturation of 0.25%. In July 2014, Leaitch et al. (2016) found a median CDNC of 10 cm<sup>-3</sup> for low-altitude clouds (cloud top below 200 m) and of 101 cm<sup>-3</sup> at higher altitudes. In October 2004, McFarguhar et al. (2007) conducted aircraft measurements in single-layer stratus clouds and found averaged cloud droplet number concentrations of 43.6±30.5 cm<sup>-3</sup>. Applying the standard CDNC threshold of 40 cm<sup>-3</sup> would drastically reduce the influence of changes in the CCN concentration and therefore impede aerosol-cloud interactions. Thus, we decided to use 10 cm<sup>-3</sup> 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 ( $1.875^{\circ} \times 1.875^{\circ}$ ), which should be less variable than a point measurement. Nevertheless, we acknowledge that the threshold of 10 cm<sup>-3</sup> 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  $\alpha$ =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 emission

"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<sub>3</sub>, CO<sub>2</sub>), which are also important forcers (Dalsøren et al., 2013; AMAP Assessment, 2015)."