Response to comments from Reviewer 1

(Reviewer's comments are copied in italic here; authors' responses are in bold)

General comments:

1. Major concern: The model runs have only been done for a short period March-October 2010 and only results for the shipping period (June –September) were presented. That is weakness of the paper, also because there are large seasonal variability in the arctic of air quality. It could be nice to see the model system performance a whole year, nice the see the overall contribution from shipping activities over a whole year, not only when shipping activities peaks. Especially in the section about deposition it is a problem, because the results covers only 25% of a whole year and there are large seasonal variability of the deposition of N and S. In line 19-20 on page 20 there are a statement that these deposition levels are in general accordance with previous estimates e.g in Hole et al, 2009, which are a whole year deposition especially in the Northern part of Canada due the large seasonal variability of concentrations, surface conditions (snow-ice-forest-tundra) and the type and amount of precipitation.

Response: We understand the reviewer's concern on the length of the model runs. As our goal of the study is to assess the impact of marine shipping emissions over the Canadian Arctic waters, it made sense to us to focus on the Arctic shipping season. The following summary table (Table R1-1) shows that there is little shipping activity outside the period between July and October over the Canadian Arctic waters. The harsh ice and weather conditions, particularly over the Canadian Arctic Archipelago, make navigation through the Canadian Arctic waters extremely difficult outside the summer months. We decided to focus our analysis on the July-August-September period as it corresponds to the busiest shipping time in the Canadian Arctic and the Arctic boundary layer is the cleanest due to inefficient transport from mid latitudes to the Arctic during this time (e.g., Sharma et al., 2004, JGR, 109, D15203). We expect that the Arctic shipping would have the largest impact on air quality in the Arctic during this time period. The main basis for the reviewer's concern on the length of the model runs in our study is with regard to depositions of S and N which have large seasonal variability. When we compared our modelled deposition levels with the studies of Hole et al. (2009) and Vet et al. (2017), we scaled up the three-month (JAS) depositions to arrive at annual deposition estimates. For the base case (i.e., 2010 with Arctic marine shipping emissions) we did extend the model run for a full year. We have now computed the annual total deposition of S and N from the full-year simulation (shown in Figure R1-1 below). Our estimate of annual total depositions of S and N (based on the full-year model simulation) are 0.5-2 kg S ha⁻¹ and 0.2-1 kg N ha⁻¹, respectively, over the Canadian sub-Arctic, and 0.1-0.5 kg S ha⁻¹ and 0.05-0.2 kg N ha⁻¹, respectively over the Canadian high-Arctic. These estimates can be directly compared with the estimates of Hole et al. (<20 to 70 mg m^{-2} , or <0.2 to 0.7 kg ha^{-1} , for both SO4 and NO3 over the eastern Arctic) and the estimates of Vet et al. (0.2-1 kg S or N ha⁻¹ over eastern sub-Arctic and 0-0.2 kg S or N ha⁻¹ over eastern high-Arctic). We have revised the discussions on S and N deposition to include the new annual results (see below under "Changes"). For assessing

the shipping emission contributions to the S and N deposition, we have also computed the ship emission contributions to the July-to-October (or JASO, 4 months) accumulated deposition fluxes of S and N at the 2010 level (in consideration of significant shipping activities in October as shown in Table R1-1 below). Results (in terms of percentage contributions) are very similar to our existing results based on the JAS period (as seen in Figure 16(b) and 16(b) in the original manuscript). Since the shipping activities in the Canadian Arctic waters are seasonal (summer), we feel that it is more meaningful to look at the shipping contributions during the shipping season.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
со	1	0	0	0	0	5	44	95	86	54	11	2
NH3	0	0	0	0	0	0	1	1	1	1	0	0
NOx	12	0	0	5	0	62	508	1,120	1,022	634	128	26
PM10	1	0	0	0	0	4	29	56	52	34	6	1
PM25	1	0	0	0	0	4	26	52	48	31	6	0
SO2	6	0	0	4	0	32	200	369	341	234	40	1
voc	0	0	0	0	0	2	18	40	36	22	5	1

Table R1-1. 2010 monthly Canadian Arctic marine shipping emissions



Figure R1-1. Annual total deposition of S (left panel) and N (right panel) based on the full-year model simulation of the base case (i.e., 2010 with Arctic marine shipping emissions)

Changes:

- Revised 2nd paragraph in section 5.2 ("On deposition of S and N") to incorporate the annual deposition estimates based on the extended full-year simulation, and added a new figure (Figure R1-1 shown above) to supplementary materials.
- Revised last paragraph in section 5.2 ("On deposition of S and N") to incorporate land-coverweighted annual deposition values from the extended full-year (2010) simulation in comparison with current critical load of acidity and nutrient N; added a new table (Table S2 in supplementary materials) showing the land-cover weighted deposition of S and N over

eastern Canadian Arctic from the full-year simulation (2010) for three periods: July-to-September (JAS), July- October (JASO), and annual (Table S2 in supplementary materials).

2. Two kind of boundary conditions are used: The MACC-IFS for the arctic boundaries and the operational GEM-MACH forecast archives for the southern boundary, because the later should better represent transport from North America. It is little confusing to use to different boundary conditions. It could be nice to see how important the use GEM-MACH for the southern boundary are for the model performance is compared to use the global MACC-IFS 3-hour resolution input (is MACC-IFS so bad for the southern boundary?). It is actual mention in the text line 28-31 page 12 that some of the over prediction in the southern part of the model domain could be related to the boundary conditions.

Response: The daily chemical lateral boundary condition fields used in the final model simulations were constructed from blending MACC-IFS chemical reanalysis for 2010 (provided by ECMWF/MACC-II at 1.25 x 1.25° resolution, interpolated to the 15-km resolution model grids) with the GEM-MACH operational forecast archives (at collocated 15-km resolution model grids; the operational GEM-MACH forecast domain overlaps a portion of the Arctic domain – see Figure R1-2 below). The consideration was that, given the better (finer) resolution used by the operational GEM-MACH (15-km) forecast in comparison to MACC-IFS reanalysis at a resolution of 1.125°, we decide to make use of the GEM-MACH operational archive as much as possible to ensure a better capture of the regional transport from U.S. northeast into our model domain, which, in our opinion, would improve our model simulation. Some initial evaluations were carried out when we were testing various chemical boundary conditions (including climatology-based boundary conditions). However we did not conduct a formal sensitivity analysis on the importance of the southern boundary condition to model performance in the Canadian Arctic and northern regions. The analysis of model performance described in section 4 indicated that the southern boundary condition mainly influenced model results at sites close to the southern boundary while it had significantly less influence on model results over central and northern Canada. A similar blending approach was used for merging the North American regional emissions (processed for the 15 km resolution model grids) with the HTAP emissions used on the portion of the domain outside the North American continent for this study.



Figure R1-2. The GEM-MACH Arctic domain (dark blue background) and the operational GEM-MACH forecast domain in 2010 (shown in lighter blue, foreground), both at 15-km resolution with collocating grids.

Specific comments:

Page 2 line 27: large part of the particular matter is SO4 and is therefore a primary emission of SO4 in the shipping source area.

Response: While it is true that a large part of particulate matters in ship plumes is particulate sulfate (SO4), most of the sulfate is formed from secondary oxidation of SO2 emitted from ship stacks. Although ship emissions do contain primary SO4, the sulfur emission from ship is mainly in the form of gaseous SO2. For completeness, in the revised manuscript, we have added particulate sulfate in the suite of ship-emitted gases and particles explicitly mentioned in the introduction.

Revised sentence: "Shipping is an important source of air pollutants. Emissions of exhaust gases and particles from ocean-going ships contain carbon dioxide (CO2), nitrogen oxides (NOx), carbon monoxide (CO), volatile organic compounds (VOC), sulfur dioxide (SO2), *particulate sulfate (SO4)*, black carbon (BC), and particulate organic matter (OM)."

Page 4 line 1: the discussion of instant dilution of ship NOx emissions in global models. It is not only due the course spatial but also coarse temporal (monthly) because of the low number of ships in the arctic. Some models (.e.g. EMEP model) have special ship_NOX tracers which do not contribute to Ozone production only to HNO3.

Response: We appreciate the reviewer's comment on other potential issues which may impact model assessment of ship emissions, in addition to the spatial resolution being discussed. Here we try to summarise the findings from existing studies suggesting that the non-linear effects associated with the unrealistic instant dilution of ship NOx emissions in global models run at coarse resolutions may affect model assessments of ship emission impact. This is to provide a context for our study and the approach we are taking in using a regional model at higher resolution. We recognise that insufficient temporal representation of ship emissions and other simplifications in some of the current models, such as the example given by the reviewer of special treatment for ship-NOx tracers in the EMEP model, may result in additional uncertainties. However, we are not aware of any existing studies

addressing these aspects, and these aspects are not the focus of our current study. Nevertheless, the issues raised by the reviewer are important and should potentially be investigated in future studies.

Page 4 line 30: is the temporal emission from shipping really hourly so you can tracking the individual ships (see also my comment above)?

Response: The base inventory is processed using the Marine Emission Inventory Tool (MEIT) based on ship movements of individual vessels tracked by the Canadian Coast Guard's tracking/logging systems, as explained in the manuscript (page 4 line 20 – 30), and the processed emissions are available at various time levels (e.g., monthly, daily, and hourly). However, as explained in the "Modelling system and simulation setup" section (under "Canadian marine shipping emissions"), for further processing to model-ready marine emissions, link-based monthly ship emissions by ship track, ship types, and fuel type were obtained from the MEIT database, along with ship route polygons and associated vessel activity information. The monthly emissions, aggregated into four ship classes, were mapped onto model grids, along ship tracks, in a form of aggregated point sources and further allocated to hourly emissions, by applying uniform temporal profiles for day-of-week and hour-of-day in the SMOKE emission processing system (http://www.cmascenter.org/smoke/). (See page 8, line 9-22, in the original submitted manuscript).

Page 5 line 26-page 6 line 18: I am missing figure (f.ex. of CO2 in order to avoid changes in emissions factors due to ECA) which shows the spatial distribution for 2030 which could be compared to 2010 of the ships emission and more information assumptions for the 2030 emissions inventory, e.g. the increase for the different ships sectors, emissions factors etc, so it is easier to compare the 2030 inventories with others.

Response: We have added CO2 numbers in Table 1 (both at 2010 and projected 2030 BAU scenario, for different ship categories). The projected increase in shipping activities in 2030 is reflected in number of trips shown in Table 1 for different ship categories/sectors. We have also added an additional plot (Figure 2(b)) of the processed model-ready Canadian marine shipping emission of NOx projected for the August 2030 (BAU) to compare with the 2010 August NOx emission shown in Figure 2(a) (previously Figure 2). The emission factors are in accordance with fuel type usage (with considerations for compliance to the current and future IMO regulations). We have added further clarification on emission factors used for the 2030 projection in the revised manuscript.

Changes:

- 1. Revised Table 1 to include emission estimates for CO2e per vessel category for 2010 and 2030 (BAU).
- 2. Revised Figure 2 to include a plot showing the model-ready NOx marine shipping emissions for August 2030 (BAU scenario).
- 3. Revised text in Section 2 to add more details on the projection for 2030 BAU scenario.

Page 17 line 14: median and maximum percentage. Is it median and maximum of the 3 months average of the individual grid points inside the sectors or is it other spatial/temporal averaged concentrations?

Response: It is the former, i.e., the mean, median, maximum are based on shipping contributions to the 3-month mean concentrations evaluated at individual grid points within a given geographical sector. We have added a statement to clarify this in the revised version.

Revised text: "Table 6 summaries the mean, median, and maximum percentage contributions from Arctic shipping emissions to the JAS average ambient concentrations of criteria pollutants for each of the 9 sectors. The percentage contributions (as defined in (1)) were evaluated at individual grid points and statistics were then computed over all grid points within a given geographical sector."