Interactive comment on "Short lived climate forcers from current shipping and petroleum activities in the Arctic" by K. Ødemark et al. Anonymous Referee #2 Received and published: 10 September 2011

We thank the reviewers for thorough and very useful comments. In response to the issues brought up, we have somewhat extended the detail level of the study and added more discussion and figures.

Especially the seasonal variations of emissions and forcing responses are now more thoroughly treated, and the discussions of uncertainties and normalized radiative forcings have been substantially revised. We hope that these revisions cover the points raised by the reviewers. Additional responses to the individual points are given inline below (in italics).

The authors present a modeling study on the impact of emissions from Arctic shipping and petroleum activities on the column amount of atmospheric aerosols and radiative forcing of climate relevant trace gases and particulate matter. The manuscript is generally well written but I have a few concerns about the method used to estimate the indirect aerosol effect and I think this deserves a more thoroughly discussion. I would also like to see a more detailed discussion in the conclusions section and a more quantitative uncertainty analysis. I suggest publishing in Atmospheric Chemistry and Physics after addressing the comments and suggestions given below.

General comments

- The indirect aerosol effect is estimated from a relation between cloud droplet number and aerosol optical depth following the work of Quaas et al. More recent studies show that this approach is not appropriate and underestimates the indirect aerosol effect. For instance Penner et al. (PNAS, in press) show that CCN do not scale with AOD: "empirical relations for ln(Nc) versus ln(AOD) derived from present-day results do not represent the atmospheric perturbation caused by the addition of anthropogenic aerosols to the pre-industrial atmosphere.". Since the Arctic is a relatively clean region, this finding should apply to this study as well. I think this should be discussed and needs to be taken into account when assessing the impact of ship emissions on RF.

Reply: The main argument in Penner et al. is that the empirical relations are for relatively small perturbations, while the change since preindustrial times is comparatively large. In the present paper, we study small perturbations related to changes in shipping and petroleum activities, and hence they should be within the validity of the empirical relations that we employ. However the reviewer's comment that the Arctic is a very clean region is valid, and rather points to the fact that we may be overestimating the indirect effect. We will make a note of this fact in the model text and in the following discussions.

The text is updated as follows:

The indirect aerosol effect is only estimated for water clouds (no mixed phase or ice clouds), nor do we attempt to include other indirect aerosol effects. While recent studies (Penner et al., 2011) have indicated that this type of estimation may underestimate the total aerosol first indirect effect over the

industrial era, it is probably better suited to minor perturbations in a clean region as simulated in the present work.

- The indirect aerosol effect is only estimated for water clouds (no mixed phase or ice clouds) (p. 21574, l. 9). It seems that in the Arctic, pure water clouds are not the dominant cloud type. I rather expect mixed phase clouds to dominate in the Arctic. This is a major point and needs to be discussed in more detail, for example, can additional clouds droplets freeze or have any other impact on the ice phase? The limitations of this approach should be named and discussed.

Reply: We agree this should be noted in the text. However we also note that there is very little litterature currently available on the indirect effect on ice or mixed phase clouds, and hence it is not easy to make good estimates here. We feel that the most honest approach is still to leave this effect untreated for a study like the present one.

- The emissions should be described in more detail. For example, what are the aerosol-size distributions used? How does the seasonal cycle look like? It also remains unclear to me from table 1 whether the emission totals are given in kt(species) or kt(S), kt(N), kt(C), etc. A figure showing the geographical distribution of, for instance, NOx and SO2 would be good. Furthermore, I think the Arctic ship emissions / emissions from petroleum activities should be put into perspective with the total (global) ship emissions.

Reply: The emission-section has been extended, and a figure of the seasonality of the shipping emissions is made. Emissions from petroleum activities is assumed to be constant throughout the year. Table 1 is updated. The figure with Nox concentration change resembles the spatial distribution of the emissions, and a note of this is now in the text, but we have not added emission-plots (other than for the seasonality in the shipping emissions).

[Myhre et al. 2007] gives all the details on size distributions, optical properties etc.

- The discussion of the uncertainties (section 4.1) is too short. The main uncertainties should be discussed in more detail and numbers should be given instead of describing everything only qualitatively.

Reply: The section is re-written, and error-bars with uncertainties are added in figure 7.

Uncertainties section re-written:

The main results from this work is given in Table 2 in terms of RF for the different components. Based on the available information it is not possible to perform a formal un-certainty propagation from emissions through CTM calculations of concentrations and radiative transfer simulations. To estimate the uncertainty in the RF numbers we therefore have to rely on estimates from other studies that often have a more global focus and make a subjective adjustment to the individual uncertainties for the Arctic region. Peters et al. (2011) do not estimate uncertainties in the emission factors for 2004 emissions. For the emissions we apply the same relative, component specific, uncertainties as were used for the shipping sector in Fuglestvedt et al. (2008). For the RF by sulphate, BC and OC aerosols we use the multimodel 1- σ range of global mean RFs from model simulation with equal emissions from the AEROCOM project (Schulz et al., 2006).

For nitrate aerosols, BC on snow and the indirect effect of aerosols we apply the range given by IPCC (Forster et al. (2007), Table 2.12), while ozone we apply the same relative uncertainties for were used inFuglestvedt et al. (2008) for the shipping sector. The uncertainties are given in Figure 9. There are some Arctic or sector specific factors that could add to the uncertainties, but is not included in the estiamtes due to lack of information. This includes potentially higher uncertainties in the activity data and emission factors for shipping and petroleum exploration at high latitudes, impcat of large variability in surface albedoes and non-linear plume effects in ship plumes (e.g. Huszar et al. (2010)).

I also would like to see the conclusions section to be extended. I am missing some explanations. For example, why is "BC in snow" so different for shipping and petroleum activities?

Reply: The emissions from petroleum activity are much higher than from shipping. They are also located on land, where they are deposited on fresh snow. The large difference is also due to the fact that shipping occur mainly during months where snow and ice-extension is at it lowest, while emissions from petroleum occur throughout the year. There is no sunlight in winter, and thus no effect on radiative forcing from black carbon on snow, but it accumulates in the snow over the winter. In spring, when the snow starts to melt, BC reappears and absorbs sunlight, affecting the radiative budget and may accelerate the melting process.

In the text:

Black carbon deposited on snow and ice alters the albedo, which accelerates the melting process in spring (Flanner et al, 2007), and can warm the surface. The radiative forcing from BC on snow is 20 mWm⁻² for petroleum activity, and 0.47 mWm-2 due to shipping (Figure 5 c) and d)). The large difference stems from different magnitude of emissions, but also in the fact that shipping occur mainly during months where snow and ice-extension is at it lowest, while emissions from petroleum occur throughout the year. There is no sunlight in winter and thus no effect on radiative forcing from black carbon on snow, however BC accumulates in the snow over the winter. In spring, when the snow starts to melt, BC reappears and absorbs sunlight, affecting the radiative budget.

If the NRF for OC is not as strong in the Artic than it is in other regions, then what about SO4? If the reason is the high surface albedo (Why? If sea ice permits shipping I presume the shipping lanes are open?) shouldn't this also be the case for the direct and indirect effect of SO4?

Reply: OC in the Arctic is not as strong as in other regions, because reflective aerosols is less "effective", in term of forcing, over bright surfaces than over dark. This argument is also the case for SO4. But for OC, this effect is stronger than for SO4 (meaning that scattering from OC (in the Arctic) is weaker than for SO4), because of high relative humidity. "The reason for stronger NRF for shipping (sulphur) in Arctic is due to the higher water uptake of aerosols since relative humidity is

high. "

In the text :

High surface albedo and lower solar irradiance reduces the NRF for emissions of scattering aerosol in the Arctic compared to global emissions, as seen for OC. For nitrate and sulphate, which are more hygroscopic than OC, the generally high relative humidity in the Arctic strengthens the NRF.

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Specific comments

- p. 21571, l.6, "... direct global warming from the GHGs": I don't like this term as I think it might be misleading. I propose to delete "direct" and simply refer to "global warming from the GHGs". *Reply: Corrected*

- p. 21571, l. 23, "... due to feedbacks occurring at these high latitudes.": This should be expanded by giving examples and adding references.

Reply: In Berntsen et al (2005) it was found that ozone perturbation caused by emissions of NOx or CO in Europe caused a larger perturbation in surface temperature all the way to the pole compared to the response to ozone perturbations cause by Asian (lower latitude) emissions. However, we agree that the RF perturbation still is is mainly located at mid-latitudes and that the results from Berntsen et al. can not be generalized to claim that RF in the Arctic cause a stronger Arctic temperature perturbation than a mid-latitude RF. There are not many simulations with climate models to simulate this, but one study (Shindell and Faluvegi, NGS, 2009) found that for greenhouse gases like CO2 and ozone there was no enhancement of the surface response in the Arctic when the RF was constrained to the Arctic compared to RF at 30-60N. For BC they even found a negative surface temperature response (for the direct effect of BC in the atmosphere). We have modified the text to make the discussion more precise.

- p. 21572, l. 21, l.25: No references are given for the OsloCTM2 and the chemistry scheme used.

Reply: References are added, and there is specifics on the point below explained.

- p. 21572, l. 23: Please be more specific which data and variables from the IFS model are used to drive OsloCTM2.

Reply: We have added references in the text. There are a number of variables used, an overview can be found at: <u>http://www.ecmwf.int/publications/manuals/libraries/tables/localTable128.html</u>, though not all are in use in the CTM. In addition, there are some fields that are not on the list:

Mass Flux updrafts Mass Flux downdrafts Mass Flux UD detrainment Mass Flux DD detrainment Precipitation (old data) Convective precipitation Largescale precipitation

- p. 21573, l. 11, "... with a more advanced BC scheme better reproducing BC measurements at high latitudes": Please give some more details here.

Reply: We have revised the BC scheme to include seasonal and latitudinal variability in the aging time (conversion from hydrophobic to hydrophilic form) based on detailed simulation with a version of the model that includes the M7 aerosol microphysics. In M7 the conversion from externally mixed BC (freshly emitted hydrophobic particles) to internally mixed BC particles (coating by sulphate only in M7) is explicitly calculated (Lund and Berntsen, ACPD, 2011). To limit CPU time needed a simplified version using look-up tables for the aging time was developed (Skeie et al., ACP, 2011). This version has now been used for this study. The effect is particularly pronounced at high latitudes where the production of sulphate is limited during winter by slow oxidation of SO2. This slows down the aging and thus the removal of BC leading to enhanced wintertime concentrations of BC in the Arctic in accordance with the observations.

This description is also added in the text.

- p. 21573, l. 12, "This scheme also includes uptake of BC on snow and ice covered surfaces.": Do you mean deposition of BC on snow/ice and corresponding changes in albedo? *Reply: Yes. The methods are described in Rypdal et al. (Tellus, 2009) and Skeie et al. (ACP, 2011)*

How is sea ice and sea ice albedo handled in the model? *Reply:* This is decribed in detail in Rypdal et al. (Tellus, 2009) and Skeie et al., ACP, 2011). Cf. text below.

Are they prescribed from observations? *Reply: No*

Is there an effect of BC on sea ice as well? *Reply : Yes*

From Rypdal et al:

A module for deposition and accumulation of BC in snow has been added to the Oslo CTM2. The module accounts for up to 10 vertical snowlayers, and total snowdepth is calculated according to snowfall, snowmelt and snowsublimation in accordancewith the meteorological input data from the ECMWF. During periods without new snowfall, dry deposition will contribute to BC in the surface layer. This is accounted for by treating a top-layer of 1 ml water equivalent separately. The effect of melting and sublimation is treated relatively simple as it is assumed to take place from the top of the snowpack, and when the whole snowlayer has melted its BC content is added to the layer below.

Thus any possible vertical movement of BC in the snowpack by drainage or sedimentation is neglected. Total BC burden in snow (including contribution from biomass burning) at the end of April is given in Fig. 5. The RF due to BC in snow is based on pre-calculated lookup tables for the relation between BC concentration in snow and ice and surface albedo. The lookup tables are calculated with a radiative transfer code (Stamnes et al., 1988) and with BC optical properties as suggested in (Bond et al., 2006) including an enhancement in the absorption to take into account internal mixture as coated BC particles. The 10 snow layers in the CTM2 model are reduced to two snow layers in the radiative transfer calculations with 1 mm water equivalent thickness of the upper layer. The calculations of the lookup tables of surface albedo are performed with 33 atmospheric layers in addition to the two snow layers. In these calculations an effective snow grain size of 500 µm is adopted. Sensitivity calculations show an uncertainty of 25% related to

the snow grain size. The BC has a larger effect on the surface albedo for larger snowgrain sizes for a given snowlayer (Flanner et al., 2007). However, larger snow grain sizes reduce the impact of the upper snow layer due to deeper penetration of the solar radiation in the snow, and the upper layer has often a higher BC concentration. Therefore, the uncertainty of the snow grain size of 25% is similar or smaller than their factors influencing this RF mechanism. Global calculations are performed with an off-line radiative transfer code for global conditions (Myhre et al., 2007).

The RF results are presented for annual means and are based on calculations with a 3-hr step. The BC impact on the snow albedo is non-linear with strongest impact at small abundances (Flanner et al., 2007). In our RF calculations we have first calculated the contribution from each region, that is, with no BC emissions from other regions. To take into account the non-linearity in the BC on snow albedo we thereafter calculated the total RF in a simulation with BC emissions from all regions included. The RF from each region was obtained by scaling the results for each region to this total.

From Skeie et al., ACP, 2011:

"The ECMWF snow depth, snow melt and snow evaporation data do not exist for grid boxes with sea ice. During winter (until mid May), snow layers are generated from the snow fall data if the sea ice extent is greater than 30 %, assuming no removal. The removal of snow during the melting season is treated quite simply. In the Northern Hemisphere, the snow is assumed to melt at a constant rate from mid May to late June. On 21 June we assume that all the snow has melted. If there is a new snowfall in this period the rate of melting is changed, so that all the snow will still be gone by 21 June. Until 8 August new snow layers on the sea ice are not allowed to form. The BC content kept at the surface of the sea ice, and the snow depth on a snow-free sea ice surface is set to 1mm of water equivalent. In the Southern-Hemisphere, snow layers on the sea ice undergo melting in the period between 15 October and 20 December, and new snow layers are not generated before 6 February. If the sea ice extent is below 30 %, all the snow is melted immediately and all the BC is removed."

-p. 21576, l. 12, "4.19 Wm-2" → "4.19 mWm-2" *Reply: corrected*

- p. 21576, l. 19, "The current methane emissions from Arctic oil and gas activity are also small in such a context. We therefore expect the radiative forcing due to direct methane emissions to be small.": Isn't this the case for the other emissions from gas and oil activities as well? Please explain.

Reply: We have now accounted for the fact that the methane emissions from petroleum is so much larger than for shipping. Our claim that the radiative forcing due to direct methane emissions from oil and gas activity is small and negligible is therefore not valid. Our calculations (method explained in comment to reviewer's next question) indeed shows that this RF is of substantial importance. The section on methane forcing are changed to include the new finding.

The section on methane forcing in the text is now as follows:

Emissions of short-lived components (CO, NOx and NMVOCs) influence the chemical loss of methane. Even if methane is not a short-lived climate forcer a significant proportion of the methane perturbation could stem from changes of short-lived components. In this section we have therefore included simplified estimates of methane RF. We used the approach described in Berntsen et al. (2005) and Myhre et al. (2011) to calculate the global radiative forcings from methane and associated ozone and stratospheric water vapor changes. The RF values from this method apply from when the perturbations have reached equilibrium conditions. Due to the relatively high NOx /CO emission ratio both the Arctic shipping and petroleum activity leads to increases in OH and thereby decreases in methane lifetime (Fuglestvedt et al., 2008). However, the changes in OH are rather small due to inactive photochemistry in winter. The effect on methane loss is further limited by low temperatures prevailing for most of the year in the Arctic. Due to this it is mainly the ship emissions that have a significant impact on methane lifetime since the traffic and effect on ozone (Figures 3 and 4) peak during summer. We find the resulting indirect methane RF (at steady state, i.e. corresponding to sustained NOx , CO and VOC emissions) from shipping to be -0.59 mWm-2 (global mean). This further results in a methane-induced ozone RF of -0.18 mWm-2 and stratospheric water vapor

RF of -0.09 mWm-2. For the Arctic petroleum activity the forcings due to changes in methane lifetime are very small amounting to -0.03 mWm-2 for methane, -0.01 mWm-2 for ozone and -0.005 mWm-2 for stratospheric H2 O. The ship emissions of methane (Table 1) are small compared to the total global methane emissions from all anthropogenic and natural sources. We find that the radiative forcing due to direct methane emissions from Arctic shipping is negligible. The methane emissions from the petroleum activity are more than a factor 1000 larger than for Arctic shipping (Table 1). The resulting change in methane concentration at equilibrium leads to radiative forcings of 1.1 mWm-2 for methane, 0.33 mWm-2 for ozone and 0.17 mWm-2 for stratospheric H2 O.

- p. 21577, l. 6, "and stratospheric water vapor RF of -0.09 mWm-2": Is a 1-year integration enough for assessing changes in the stratosphere? Because of the longer time scales involved, the stratosphere might not have reached equilibrium yet and the numbers are thus not representative. Please explain.

Reply: The method for calculating the forcing due to CH4 changes follows the approach described by Berntsen et al. (2005) and Myhre et al. (2011). We avoid repeating the details of the method in the text but instead refer to those studies. However, it is now noted in the text that the methane RF, CH4-induced O3 RF and stratospheric H2O RF apply for equilibrium conditions.

The CTM do not explicitly calculate the change in CH4 concentrations and in any case the simulations are not long enough for the CH4 to come into equilibrium with the changed OH field. Instead, the OH imposed fractional change in CH4 lifetime calculated by the CTM is used. For the effect from direct methane emissions the fractional contribution to total global emissions are used. The fractions are then multiplied by the present-day concentration of methane and a model-average feedback factor of 1.4

(IPCC 2001), to account for the impact of changes in CH4 concentration on its own lifetime, to yield the fractional changes in CH4 concentration for steady-state conditions. The indirect (through OH) and direct RF is calculated assuming a specific CH4 RF of 0.37 mW m²/ppbv (IPCC 2001), which assumes a background concentration of 1740 ppb for methane and 319 ppbv for nitrous oxide (IPCC 2001).

The radiative forcing due to the methane-induced ozone change is estimated using multimodel means (IPCC 2001) of both the response of ozone to a methane change (a 10% increase in methane leads to a 0.64 DU increase in ozone) and an ozone specific radiative forcing of 42 mWm² /DU. The stratospheric water vapour RF is set to be 0.15 times that of the CH4 RF. (See Myhre et al. 2011 for reference)

In the above calculations it is assumed that the CH4 concentration in 2004 is in steady state with that year's change in OH. The actual degree of imbalance depends on the history of change in OH, which is not accounted for in the chemical model calculations which used year 2004 emissions. The degree of imbalance will be largest for emission sectors like shipping growing rapidly in recent years. The factor to correct this transient response in year 2004 is taken from Myhre et al. 2011 to be 0.8 for the shipping sector. For the petroleum sector we assume similar change as the total global methane emissions change and therefore set the factor to 1. These factors are then applied to the CH4 RF, the CH4-induced O3 RF and the stratospheric water vapour RF.

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- p. 21578, l. 10, "... the radiative balance from reflective aerosols over bright surfaces is small.": What about sea ice and changes in sea ice coverage that make shipping in this region possible in the first place? At least the open shipping lanes should have a low albedo. Please comment.

Reply: Even if the open shipping lanes have low albedo the area is currently dominated by bright surfaces with snow and ice for much of the year. We think this explains the weaker OC NRF from shipping compared to former global studies. Since OC has a lifetime of a few days it is relatively effectively transported out of the shipping lanes towards areas covered by snow and ice. In contrast to some other components there are no chemical processes in the ship plume shortening its lifetime making the OC perturbation limited to the shipping lanes.

Change in the sea ice-extent due to a warmer Arctic with the changes in shipping patterns this brings with it, will be presented in a different paper, with emission-inventory for future emissions. This paper focuses only on current emissions and conditions.

- p. 21579, l. 1, "The indirect aerosol effect dominates shipping contribution, ...": What about changes in water vapor (as a results of changes in cloud properties) and CO2 (even though CO2 is not short-lived). It would be helpful to put things into perspective. It should be stressed that this statement takes only short-lived species into account.

Reply: An indirect effect of aerosols through clouds and thus water vapor is a very interesting issue, but currently there has been very little research into this topic and we judge that it would be premature to put any numbers on the possible magnitude of this effect. In a paper by Pfeffer et al. (just submitted to ACPD) we have started to investigate this effect.

However, in general one would believe that this effect would be more important at lower latitudes where deep convection is a major source of water vapor to the free troposphere (where the greenhouse effect of WV is stronger due to the higher temperature contrast with the surface).

The GWPs for the short-lived components given in Table 4 can be used to make first-order assessments of the global impact of changing emissions from the shipping and petroleum sectors in the Arctic. The net impact in terms of changes in "CO2 -equivalent" emissions for any given scenario, including LLGHGs with GWPs from IPCC (2007), can be derived by calculating the sum of the GWP-weighted emissions for all components.

M. A. Pfeffer, J. E. Kristjansson, F. Stordal, T. Berntsen, and J. Fast, Indirect radiative forcing of aerosols via water vapor above non-precipitating maritime cumulus clouds, Atmos. Chem. Phys. Discuss., 11, 27637-27659, 2011).

- p. 21580, l. 7: Add "of aerosols" after "water uptake" *Reply: corrected.*

- p. 21580, l. 7, "Sulphate from petroleum are...": "are" \rightarrow "is" *Reply: corrected.*

- p. 21581, l. 14, "Sulphate values are about 2 times stronger in Fuglestvedt et al. (2010) than here, due to the prevailing Arctic conditions.": This should be explained in more detail and might not necessarily be true as the method applied to estimate the indirect aerosol effect might simply underestimate the aerosol effect (see general comments).

Reply: We agree and will add more discussion here, though see our response to the general comment above. More discussion on effects regarding sulphate is added several places in the text.

- p. 21582, l. 7: "leads" \rightarrow "lead" *Reply: corrected*.

- p. 21582, l. 9: "exert" \rightarrow "exerts" *Reply: corrected.*

- p. 21582, l. 19: insert "a" before "different" *Reply: corrected.*

- p. 21582, l. 21: "show" \rightarrow "shows" *Reply: corrected.*

- p. 21582, l. 26: "are stronger" \rightarrow e.g. "has a stronger GWP" *Reply: corrected.*

- p. 21583, l. 1: "GWP" \rightarrow "GWPs" *Reply: corrected.*

- p. 21589, caption of table 1: Insert "in the Arctic" after "activity" *Reply: corrected.*

- p. 21590, table 2: The values for "BC snow" are missing. I presume this would be the amount of BC deposited on snow in the year 2004? *Reply: The table shows the atmospheric columns, BC in snow is not included, because it is not a atmospheric column value. However we've included it in the same table to display the RF values.*

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- p. 21592, caption of table 4, "Sulphate are" \rightarrow "Sulphate is". *Reply: corrected.*

- p. 21592, table 4: Are the emissions assumed to be constant over the years? Please clarify. *Reply: New plot added, and text updated.*

- p. 21597, figure 5: Shouldn't the lower limit of the color scales be zero? *Reply: corrected.*

- p. 21598, figure 6: Is this the change in net cloud forcing or in shortwave cloud forcing only? *Reply: SW only.*