Reply to the review of "Aerosol indirect effects from shipping emissions: Sensitivity studies with the global aerosol-climate model ECHAM-HAM" (acp-2012-139) by Jeff Pierce

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This paper explores the effect of ship emissions on aerosol microphysics and the climate effects of these aerosols. Experiments were performed to test the sensitivity of the simulation results to uncertain inputs such as the total mass emission rates, particle size, hygroscopicity and composition. The simulated radiative forcings depend greatly on the assumed inputs and highlight the need for continued research on ship emissions.
The paper is generally well written and well within the scope of ACP. I recommend this paper be published once the following comments have been addressed.

Thank you very much for this concise summary of our work and we very much appreciate Jeff Pierce's recommendation of publishing our paper after we have adequately addressed his comments.

There may be considerable uncertainty in the AIE forcings due to the prediction of clouds in ECHAM-HAM and the predicted susceptibility of cloud changes to aerosol changes. Two models with identical aerosol emissions may have very different AIE predictions due to ship emissions. This should be discussed.

Thank you very much for this indeed important comment. The AIE as obtained from GCM simulations depends on a number of factors, of which the GCM-simulated cloud fields and cloud microphysics are most probably of highest importance. For the specific case of ECHAM5-HAM, we will discuss

these points separately:

Cloud cover

For evaluation of the models' simulated cloud fields, it appears useful to compare them to observations, e.g. satellite data. This cannot be performed straight forward, but requires the application of so-called "satellite-simulators" to the model output to ensure direct comparability of modelled and observed cloud fields (e.g. Bodas-Salcedo et al., 2011). For the case of the ECHAM5 model, this has just recently been performed in the studies of Nam and Quaas (2012, in press) and Gehlot and Quaas (2012, in press), where Nam and Quaas (2012, in press) compared the ECHAM5-simulated cloud fields to CALIPSO and CloudSat observations whereas Gehlot and Quaas (2012, in press) used ISCCP observations for the same purpose. In both studies, the convection parameterisation is based on Tiedtke (1989) with modifications for penetrative deep-convection according to Nordeng (1994), which is the same setup as for our simulations. However, in the simulations of Nam and Quaas (2012, in press) and Gehlot and Quaas (2012, in press), cloud cover is calculated by employing the scheme of Tompkins (2002) whereas we employed the cloud cover scheme of Sundqvist et al. (1989). Our experience shows that the two cloud schemes produce quite similar cloud distributions in a present-day climate. We are thus confident that the results presented in Nam and Quaas (2012, in press) and Gehlot and Quaas (2012, in press) are also applicable to our simulations.

Both of those studies found that ECHAM5 overestimates high-cloud cover and underestimates mid- and low-level cloud cover. The overestimation of high cloud cover is attributed to the convective scheme transporting too much water into the upper troposphere by not allowing for enough detrainment at lower levels. This then leads to too many, too thick and too high clouds, especially in the tropics where convection is intense, at the expense of mid- and low-level clouds. If simulated, often a too low coverage by low-level clouds is simulated (Nam and Quaas, 2012, in press). From common sense, one expects low-level liquid water clouds to be most susceptible to shipping emissions. The underestimation of such clouds in ECHAM5 may in fact explain the noisiness of the AIE signal in the tropics. For the mid-latitudes, i.e. where the bulk of shipping emissions occurs, low-level clouds seem to be represented reasonably well.

Overall, this deficiency of the ECHAM5 model in correctly representing low-level liquid water clouds may lead to an underestimation of calculated AIEs. We have added some discussion of this into the manuscript (the "model description" section.):

"Gehlot and Quaas (2012, in press) and Nam and Quaas (2012, in press), using satellite observations, evaluated the ECHAM-simulated cloud cover using the same convection parameterisation but a different cloud cover scheme, *i.e.* that of Tompkins (2002). Both studies revealed that this model configuration overestimates high-cloud cover at the expense of mid- and low-level cloud cover, especially in the tropics and subtropics. As the ECHAM-simulated cloud fields are similar for both cloud-cover schemes (Quaas, 2012), these findings also hold for the model configuration we use in this study. As shipping emissions are most probably bound to impact the properties of low-level clouds, the AIEs obtained with this model may represent a low estimate, especially for tropical and subtropical regions."

Cloud microphysics

Whereas the cloud fraction distribution is mainly determined by the processes described above, the susceptibility of cloud changes to aerosol changes is (in the model environment) determined by the employed cloud microphysics scheme. By using the double-moment scheme of Lohmann et al. (2007), non-linearities in the cloud-aerosol system are accounted for. Indeed, this yields a substantial advantage over models using single-moment cloud microphysics due to the simplistic view of aerosol-cloud interactions in such parameterisations.

Another important factor is the parameterisation of autoconversion, i.e. the conversion of cloud water to precipitable water. In the model version used here, this is performed via the empirical relationship proposed by Khairoutdinov and Kogan (2000) where the autoconversion rate correlates negatively with the number of cloud droplets. Thereby, cloud-lifetime effects are explicitly "hard-coded" into the model and any results regarding such effects should be handled with care. We now comment on the autoconversion scheme in more detail:

"Autoconversion, *i.e.* conversion from cloud droplets to form precipitation, is treated according to Khairoutdinov and Kogan (2000), an empirical relationship where autoconversion negatively correlates with the number of cloud droplets. Thereby, cloud lifetime effects are explicitly included in the model. It is thus likely that the model overestimates the "cloud lifetime effect" (Quaas et al., 2009)."

We have also inserted the following note to the very end of the revised manuscript:

"As there exists a considerable inter-model spread regarding total AIEs

(*e.g.* Penner et al., 2006; Quaas et al., 2009), it must be noted that the estimated range of AIEs stemming from shipping emissions crucially depends on the employed GCM model setup, encompassing the used convection, cloud cover and aerosol- and cloud-microphysical parameterisations. Future effort should therefore focus on performing model intercomparison studies of not just total aerosol indirect effects, but also of aerosol indirect effects attributable to a certain economical sector, *e.g.* shipping emissions."

Observable ship tracks generally have widths that are smaller than the grid resolution used here. Given that the shift between open-celled and closed-celled straticumulus clouds may be non-linear with aerosol concentration, there may be uncertainties in the predicted AIE due to these plume-cloud effects. This should be discussed.

We very well acknowledge that i) mesoscale dynamical features cannot be resolved in current global modelling approaches and ii) simulating the effect of shipping emissions on clouds by using constant emission fluxes over a whole grid box (leading to substantial dilution of the emissions) is not quite realistic. However, we are limited by the spatial resolution of the employed emission inventory (1x1 degree). Confining the shipping emissions to less wide corridors and accounting for sub-grid processes could indeed lead to different results.
Previously, Franke et al. (2008) assessed the impact of employing a sub-grid plume model to shipping emissions in comparison to instant dilution into a GCM-scale grid-box. They focused their analysis on ozone chemistry and found that accounting for sub-grid scale effects does indeed have important impacts on the results. They propose employing "effective emissions" in the GCM environment, i.e. emissions modified by the effect of unresolved processes in the model. Unfortunately, Franke et al. (2008) only present results obtained from a box-model, implementation into a GCM was, to our knowledge, not performed. An assessment of the potential effect on cloud micro- and macro-physics is not performed.
Employing such an approach of "effective emissions" is indeed very attractive, but well beyond the scope of this study. A nice review of the possibilities is given in Paoli et al. (2011). We have added two passages on these issues to the description of the model setup:

"In addition, mesoscale dynamical features, such as the transition from open-cell to closed-cell stratocumulus clouds (*e.g.* Wang and Feingold, 2009; Sandu et al., 2010) cannot be represented in current global modelling approaches."

"We acknowledge that applying the emissions in such a way leads to substantial dilution of the emissions, thereby neglecting sub-grid processes of emission processing in the atmosphere (*e.g.* Stevens et al., 2012). The possibility of employing effective emissions (Paoli et al., 2011), *i.e.* emissions modified to represent the effect of unresolved processes on the scale of a GCM grid-box, was previously explored by Franke et al. (2008). In that study, the authors found substantial effects on ozone chemistry, but neither did they discuss possible implications for AIEs nor was the method applied to a full GCM. More recently, Huszar et al. (2010) employed an exhaust plume parameterisation for shipping emissions on a regional scale (Eastern Atlantic and Western Europe) using a model environment encompassing a regional climate model and a chemical transport model. Similar to Franke et al. (2008), they found substantial effects on atmospheric chemistry, mainly comprising reduced abundance of NO_X and ozone over main shipping corridors compared to a simulation utilising the standard emission implementation of instantaneous dilution. Although these results are stimulating, including sub-grid scale effects on emission processing is well beyond the scope of this study and we therefore stick to the conventional method of instantly diluting the shipping emissions to a GCM grid-box. "

The subgrid nucleation and growth in sulfur-rich plumes (relevant to the number and size of the sulfuric acid particles from ship emissions) depends greatly on the atmospheric conditions. Obviously day/night, sunny/cloudy conditions will affect the nucleation and growth. Additionally, if the background air already contains a lot of aerosols, nucleation/growth will be strongly suppressed as nucleation will be slower, growth will be slower and coagulation will be faster. Thus, more particles should be formed in cleaner environments and fewer in polluted. This is explored in detail for coal-fired power plants in"Stevens, R. G., Pierce, J. R., Brock, C. A., Reed, M. K., Crawford, J. H., Holloway, J. S., Ryerson, T. B., Huey, L. G., and Nowak, J. B.: Nucleation and growth of sulfate aerosol in coal-fired power plant plumes: sensitivity to background aerosol and meteorology, Atmos. Chem. Phys., 12, 189-206, doi:10.5194/acp-12-189- 2012, 2012", but the results should be general for ship emissions too. This dependence of sub-grid nucleation/growth on background aerosol could affect the size distribution and forcing predictions, and this should be discussed when presenting the assumed emission size distributions of sulfate.

Thank you for this important remark. On the scale of a model time step, aerosol processing in HAM accounts for the mechanisms mentioned in this

comment. Therefore, the processes simulated in HAM should in principle (on a large scale and neglecting plume effects) represent those mentioned in Stevens et al. (2012), who observed the evolution of the aerosol population inside a plume for several hours.
As this comment basically covers the same issue as raised in the previous comment, we do not add anything specific to the manuscript at this point but add a reference to Stevens et al. (2012) (see above).

What is the definition of statistical significance here throughout? Is this that there is a <10% of a chance that the difference between the two simulations (averaged over 5 years) could have happened randomly? Perhaps this was specifically stated and I missed it, but if not, please add.

Yes, this is exactly the way the statistical significance is defined. We have added the following phrase to the manuscript:

"The level of statistical significance applied to all plots is 90%, *i.e.* the null hypothesis (that the sample means are from the same population) is true with <10% probability and is thereby rejected in the regions indicated by the contours in the plots. "

How does nudging affect cloud susceptibility? I could imagine that nudging could prevent clouds from fully responding to aerosols. Has this been tested before? Please comment on this.

Thank you very much for this important remark and there are indeed implications related to cloud formation. Because the nudging is performed for just four parameters (vorticity, divergence, temperature and surface pressure), the hydrological cycle remains almost untouched so clouds can develop freely within the imposed dynamics. However, a possible feedback of changes in cloud properties on the general circulation cannot be obtained from this kind of simulation and the energy budget may not be closed on local scales (S. Rast, pers. comm., 2011). However, nudged simulations are the tool of choice in this study because this allows for the derivation of statistically significant results from relatively short simulations (i.e. five years in this case). Otherwise, longer simulations would be necessary to eliminate internal variability of the model.
We have in fact not tested the effect of running a "free" simulation (mainly due to time constraints and limited computational resources), but it has previously been shown that AIEs obtained from nudged simulations compare very well to those obtained from free simulations (Lohmann et al., 2010).

We have added the following phrases to the manuscript:

"A possible feedback of changes in cloud properties on the general circulation cannot be obtained from this kind of simulation. However, nudged simulations are the tool of choice in this study (as for many other AIE studies too) because this allows for the derivation of statistically significant results from relatively short simulations (*i.e.* five years in this case). Furthermore, Lohmann et al. (2010) have shown that AIEs obtained from short nudged simulations compare very well with those obtained from "free" simulations."

Specific comments

P7077 L27-28: Please give a quick description or estimation of why reduced AIEs occurred despite increasing fuel consumption? Is this because of improved emissions controls?

In the Lauer et al. (2009) study, the authors tested the implications of emission control areas on the obtained AIEs. Their control simulation, representative of the year 2002, used a global fuel sulfur content of 2.7%. Sensitivity experiments were conducted for emissions representative of the year 2012 where total emission/fuel consumption was increased by a uniform global annual growth rate of 4.1%. The 2012 sensitivity emission inventories were designed in such a way as to mimic the use of fuel with reduced sulfur content (0.5% or 0.1%) within 200 nautical miles of any coastline. A scenario using a globally reduced fuel sulfur content of 0.5% for the year 2012 was also performed. Emissions of particulate organic matter were also reduced in the emission control scenarios.
In detail, the resultant globally averaged AIEs obtained from the year 2012 simulations mimicking emission control areas near coasts are just slightly larger than the year 2002 emissions using lower total emissions. However, the 2012 scenario in which the fuel sulfur content is reduced to 0.5% on a global scale yields a substantially lower ToA forcing compared to the 2002 control simulation. So indeed, the reduced AIEs can be attributed to the effect of emission controls.
We have added the following short phrase to the introduction:

"In their simulations, a reduction of the globally averaged fuel sulfur content from 2.7% (in 2002) to 0.5% (in 2012) lead to a reduction of globally averaged AIEs from shipping emissions from about -0.43 to about -0.27 Wm^{-2} despite an annual growth rate of fuel consumption of 4.1%. "

P7078 L11-12: You suggest combustion of cleaner ship fuel leads to reduced emissions of BC, reducing potential cooling effect – but it would also reducing BC warming as stated on previous page. Correct?

Yes, this is correct. We have modified the sentence so that we also account for that effect:

"Furthermore, evidence suggests that combustion of cleaner ship fuel also leads to reduced emission of particulate BC (Lack and Corbett, 2012), thereby reducing its potential cooling and warming effects. We here present an initial investigation of the magnitude of these effects. "

P7079 L17: In Kazil et al. (2011), they used activation nucleation in the continental boundary layer (not in the marine boundary layer), and the ion-induced nucleation every else, correct? This is an important detail and should be mentioned.

Yes. Neutral and charged nucleation is used throughout the troposphere whereas activation nucleation is limited to the forested boundary layer. We have added this to the model description.

P7081 L7: COADS should be Comprehensive Ocean-Atmosphere Data Set.

Thank you for hinting at this miswording. We have modified it in the revised manuscript.

P7081 L23: Do emissions inventories include days at port? Are these emissions substantial? If so, how does the model handle this emission input and uncertainty?

Thank you very much for this remark. In fact, the Behrens (2006) emission inventory gives information regarding fuel consumption in ports. These are estimated at 11.6 Mt/year, thus being about 6.3% of total fuel consumption. However, these emissions are not included in the spatial inventory used in our simulations. We have added a note on this to the revised manuscript:

"The spatially gridded QUANTIFY inventory we use in this study does not include emissions in ports. Fuel consumption in ports is estimated at 11.6 Mt for the year 2000, thereby representing about 6.3% of total fuel consumption (Behrens, 2006). "

Section 2.4.: Please discuss the NS experiment. I deduced from the Table that this experiment is the same as the other experiments in every way except that all ship emissions were turned off; however, this never mentioned in the text. The first use of "NS" in the text comes later, but not in reference to the simulation, but to the soluble nucleation mode. The NS simulation is eventually mentioned later, but not defined. The two uses of "NS" is a bit confusing.

Thank you very much for pointing this out ! We have now renamed the NS experiment to CTRL due to the confusion that the multiple use of NS may cause. We have also included a short description of the reference experiment in Section 2.4.:

"For our reference simulation **CTRL**, we run the model with the emissions prescribed for the AeroCom setup (Dentener et al., 2006) excluding all shipping emissions. This simulation thus yields the base case scenario for estimating the AIEs from shipping emissions as computed from the sensitivity experiments described below."

Section 2.4: I think that the experiments testing the reduction of BC/OC are very interesting as they are relevant to the recent discussion of climate controls through soot reduction. However, many regions of the world are beginning to require ships to use fuels with lowered sulfur content. It would be interesting to test this effect.

Yes, this would indeed be an interesting effect to study. However, lesser fuel sulfur content is found to generally lead to cleaner fuel combustion and thus less emissions of BC/OC (see Lack and Corbett (2012) for a recent review). Compiling a representative emission inventory for assessing the effect of fuel sulfur content reductions near coastlines (see *e.g.* Lauer et al., 2009) or even on a global scale is thus beyond the scope of this study and should be left to future modelling efforts.
We have added the following note to the revised manuscript:

"Although very intriguing, we explicitly do not attempt to assess the climate impact of emission controls resulting from a reduction of fuel sulfur content on local and global scales (IMO, 1998; Lauer et al., 2009; Righi et al., 2011) and this should thus be an important topic of future climate model intercomparison studies."

Section 2.4 and throughout: I find the use of "emissions parameteriza-

tion" a bit uninformative. It would be more informative to say emissions size and composition (a bit tricky since it also includes the fraction of SO2 oxidized on the sub-grid scale).

Thank you for this comment but we prefer to stick to the naming convention that we have adopted in the submitted manuscript. We do so because we think that our wording is sufficiently exact. Generally, the term "parameterisation" refers to a formulation which represents the effect of unresolved processes on large-scale resolved variables. This formulation then can include a large number of individual effects, which for the case of our term "emission parameterisation" includes the representation of emission size distribution and composition as well as fast processes. The term "emission parameterisation" therefore meaningfully summarises all assumption in one term and is thus also of advantage for the readability of the paper.

Section 2.4: Related to the previous comment, it would be useful to know how much of the differences between the A and B experiments is due to the difference in size of sub-grid sulfate, the fraction of SO2 oxidized in the sub-grid scale or the hygroscopicity of the emissions. If short simulations could be ran to give global-average BL estimates of what each of these factors contribute, this would be very helpful in determining the most important parameters to measure/improve.

Yes indeed, that would be interesting to investigate. Following Jeff Pierce's suggestion, we performed additional short experiments, each for 1 year only (2000, simulations include a three-month spin-up period of Oct-Dec 1999), to find out which of the changes contributes most to the differences between experiments **A** and **B**. The setup of the additional experiments as well as the resulting AIEs for that particular year are shown in Tab. 1.

From these experiments it is evident that it is the change in the emitted particle size distribution which has the largest effect on the resulting AIEs. Combining the increase of $f_{\mathrm{SO_4}}$ with a smaller emission size distribution (S4) yields AIEs very close to those of **B**. The decrease of the AIE in experiment S1 with respect to **A** is at first surprising. We do not analyse that simulation in detail here, but this result could be partially explained by a shift of the aerosol size distribution away from its optimum for aerosol activation (to larger sizes), which can then also lead to a reduction of CDNC although the emitted aerosol mass increased (*e.g.* Korhonen et al., 2010; Pringle et al., 2012). The linear superposition of the AIEs resulting from changing either one of the three factors does not yield the AIE-estimate of **B**, but it is rather the convolution of all effects and the non-linear nature of the aerosol system

Experiment	f_{SO_4} [%]	sulfate as KS	BC/OC soluble	AIE [Wm^{-2}]
A	2.5	no	no	-0.07
B	4.5	yes	yes	-0.25
S1	4.5	no	no	-0.06
S2	2.5	yes	no	-0.14
S3	2.5	no	yes	-0.08
S4	4.5	yes	no	-0.20

Table 1: Setup of the sensitivity experiments and the resulting AIE for a 1 year simulation (2000). The corresponding values for experiments **A** and **B** are shown for reference.

which then yield the AIE-estimate of **B**.
We added some information on this into the revised manuscript:

"We performed a preliminary investigation to pinpoint the process responsible for most of the increase in radiative forcing. In our study, the increase in the number of emitted soluble particles in experiment **B** compared to **A** is caused by three factors: i) increasing f_{SO_4}, ii) changing the emission size distribution of primary sulfate particles and iii) assigning carbonaceous emissions to the soluble instead of the insoluble Aitken mode. We find that it is the change in the emission size distribution of primary sulfate particles which makes up for the bulk of the change in AIE. Future measurement campaigns should thus focus on characterising the evolution of the emitted particle size distribution from the scale of the emission plume to the scale of global model resolutions."

P7083 L6: I never understood how new coarse-mode aerosol could EVER be formed through sub-grid nucleation and growth. There really is no justification for this. The use of this aspect of the AERCOM inventory really needs to stop. I'm glad that the authors are using something different for the base-case scenario. This should be extended for all sub-grid sulfur sources too in the future.

Thank you very much indeed for this affirmation of our approach in this matter. But in fact, the emitted coarse mode particles from industrial sources in AEROCOM were thought to represent fly ash rather than particles formed from sub-grid nucleation and growth (Dentener et al., 2006). However, as this is not confirmed from measurements of shipping emissions we plan to definitely feed our setup back to the next model versions.

P7083 L11-16: The fraction of SO2 oxidized to H2SO4 should depend greatly on the time from the source where the measurement was taken (not to mention the amount of sunlight and NOx and VOC concentrations that determine the OH concentration that determine how quickly SO2 is oxidized). How does the time since emission (and other factors) differ between estimates, and how does it differ from these factors in the model? Also, its not obvious to me why high-sulfur vs. low-sulfur fuels should have a different fraction oxidized as SO2 concentration does not greatly affect OH concentration. Perhaps NOx or VOC concentrations also change between the fuel types? Or perhaps there were not enough measurements to be confident in the differences between low and high SO2 fuels. (This final issue with high-sulfur vs. low-sulfur fuel is somewhat tangential to your paper, but it might be worth mentioning if you have any insight.).

In the model, oxidation of the fraction of primary emissions (e.g. the 2.5% of SO_2 emissions normally attributed to particulate sulfate at the point of emission) happens instantly at the point of emission, i.e. within one model time step (about 15 minutes in this case). For the already present SO_2 in the model, oxidation happens on a scale corresponding to the e-folding lifetime of SO_2. Aqueous oxidation may speed this process up (see next comment). In the following, we present some detail on the measurement setup in the studies cited in the manuscript:

- Lack et al. (2009): measurements of a research vessel intersecting emission plumes up to 15 minutes after emission from the stack, *i.e.* comparable to the model time step used in our study. However, they do not give any information on why the fraction of SO_2 oxidised to sulfate seems to depend on the fuel sulfur content. Their measurements did not yield any substantial differences of NO_3 measured in the plumes.

- Agrawal et al. (2008) appear to have measured the exhaust gas in the stack almost directly at the point where the exhaust leaves the engine. This implies a substantially shorter timescale than used in the model time step. They find that the fraction of SO_2 directly oxidised to sulfate also depends on the engine load, with higher fractions obtained for higher engine load.

- In another study (now also cited in the manuscript), Agrawal et al. (2010) also found fractions of 2.4 - 5% when sampling the same engine as in their 2008 paper (above). The results are again found to be dependent on engine load.

We have now added parts of this information to the revised manuscript:

"First, recent studies indicate that the f_{SO_4} is often larger than 2.5%. Agrawal et al. (2008) performed exhaust gas measurements inside a containerships' stack and found a value of $f_{SO_4} = 3.7 - 5\%$ (positively correlated with engine load) for a fuel sulphur content of 2.05%. These results are confirmed by Agrawal et al. (2010) ($f_{SO_4} = 2.4 - 5\%$ with fuel sulfur content of 3.01%). Lack et al. (2009) measured ship-emission plume compositions within 15 minutes post-emission and deduced $f_{SO_4} = 1.4\% \pm 1.1\%$ and $f_{SO_4} = 3.9\% \pm 2.0\%$ for low (<0.5%) and high (>0.5%) fuel sulphur content, respectively. The authors however do not give information on the dependence of f_{SO_4} on the fuel sulfur content. Applying the current state of knowledge, we increase f_{SO_4} from 2.5% to 4.5% in experiment **B**, being a realistic estimate considering that the model time step is on the order of the measurement timescale of Lack et al. (2009) and that the globally weighted marine fuel sulphur content is estimated at 2.68% for 2002 (Endresen et al., 2004) . "

P7085 L24: Aqueous oxidation is generally estimated to be somewhat more important for taking SO2 to sulfate globally, and this would depend on the cloud cover amount as well as H2O2 and O3 concentrations (though these concentrations probably correlate well with OH).

Thank you very much for this important comment. We now also mention this process in the revised manuscript:

"Also, aqueous oxidation in cloudy scenes is most probably higher over the North Atlantic compared to Southeast Asia due to higher cloud fractions in low- and mid-levels (Nam, 2011). "

P7087 L3: The second explanation in this sentence is incorrect. The additional particles emitted to KS would increase the condensation sink and would lower [H2SO4] for a constant H2SO4 production rate. Lower [H2SO4] would LOWER the growth rate of the Aitken-mode particles to accumulation-mode sizes, not make them faster. The likely reason for the faster growth of Aitken-mode particles to accumulation-mode sizes is the additional SO2 emitted by the ships that would increase [H2SO4].

Thank you very much for pointing this out. After considering Jeff Pierce's line of argument here, we agree with him and changed the manuscript accordingly:

"The cause for this is two-fold. Firstly, less nucleated NS-particles (see above) result in less particles growing to KS and secondly, the increased availability of condensable material (H_2SO_4) leads to higher condensation rates onto the already present and additionally emitted KS particles."

P7088 L2: Increasing the size of particles from the Aitken-mode to the accumulation mode would decrease dry-deposition rates, not increase them (dry deposition rates have a minima in the accumulation mode). However, the wet-deposition would increase due to more particles being large enough to act as CCN, and this effect likely dominates over the lowering of the dry-deposition rates.

Thank you very much for this comment. We agree and apologise for this miswording. We have now changed the manuscript accordingly:

"The aforementioned faster growth rates of aerosol particles could in fact lead to a reduction of their atmospheric lifetimes due to enhanced wet deposition through cloud interaction."

P7088 L7 and the caption of figure 5: AOD does not have a fine mode. You could say "total AOD and the fine-mode AOD" or "AOD and its fine-mode component", but "the AOD and its fine mode" seems awkward to me.

Yes indeed. Thank you very much for pointing at this miswording. We have adopted the manuscript accordingly.

P7089 L10: Why is there so much noise in the cloud property changes even when averaged over 5 years? The magnitude of the noise is very large.

This is indeed a very interesting point and to this point, the only explanation we can give is the one already given in the manuscript. Over tropical oceans, cloud formation is most likely not dominated by large-scale stratification but by convective processes. As shown in the work of Gehlot (2010); Nam (2011); Nam and Quaas (2012, in press) and Gehlot and Quaas (2012, in press), the convection scheme used in ECHAM5 substantially underestimates low- and middle level cloud cover due to too less detrainment of water vapour at those levels. This then leads to an overestimation of high (ice) clouds. Therefore, the variability of low- and mid-level cloud microphysical properties is too large to give statistically significant results. On the other hand, this lack of significance over tropical oceans is supported by our previous study (Peters et al., 2011), namely that natural variability in these

regions is too large to allow for a signal-to-noise ratio sufficient to indicate a statistically sound microphysical effect. We have added reference to our work in the revised manuscript:

"This is in-line with the findings of Peters et al. (2011) and a more thorough comparison to observations will be performed in the future. "

P7091 L8: I would have guessed that a cloud thickening would cause a reduction of outgoing longwave radiation. Higher cloud-top heights have colder temperatures and would emit less longwave radiation.

Thank you very much for pointing this out. Indeed, this behaviour would be expected according to previous work on LW aerosol indirect effects (Koren et al., 2005; Devasthale et al., 2005; Teller and Levin, 2006). To investigate this, we checked the postprocessing routines again and found that everything was computed correctly. Due to the fact that these results show considerable uncertainty, we removed this sentence and the plot showing the change in OLR in the revised manuscript to avoid any confusion.

P7092 L1-6 and Fig 9: species column burden differences between experiment A & B in the specified hemispheres are difficult (and at times not able) to discern.

Thank you very much for hinting at this problem in readability of our results. We have now modified the plots in a way to ensure readability: for the SO_2 column burdens, we only show changes for experiments B and Bsc because the other experiments are very similar to either one of the curves. Regarding the changes in sulfate column burdens, we show results for experiments A, Asc, B and Bsc only. The results for BnoC and BnoBC are similar to B and are thus not shown. Furthermore, we also enhanced the font size of the legend of all zonal plots in order to increase readability. We also modified the respective plot caption:

"Relative changes in SO_2 column burdens obtained from experiments **A**, **BnoC** and **BnoBC** are very similar to **B** whereas changes in **Asc** are very similar to those in **Bsc**. Changes in sulfate column burdens in **BnoBC** and **BnoC** are very similar to those obtained from **B**."

Section 4.1: Please explore/discuss the contribution to the differences between A and B from sulfate aerosol size, solubility and the fraction of SO2 oxidized on the sub-grid scale.

Please see our discussion above.

P7094 L3: inrease = increase

Thank you, we have corrected this in the revised manuscript.

P7097 L28-: SO2 mass emissions from ships are nearly 2 orders of magnitude larger than OM and BC emissions. This means that the ship OM and BC emissions are located in regions where the potential for nucleation is elevated (at least compared to if the SO2 emissions were missing). This makes the OM and BC from ships somewhat unique compared to many other sources of OM and BC where the SO2 emissions are smaller.

Yes indeed and that is also what we already mention in the paper (although in shorter form). We have added some information to our previous phrasing of this effect and this passage now reads:

"This effect may however not appear in regions in which the potential for new particle formation is lower, *e.g.* already heavily polluted industrial areas. "

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