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Dr. Galmarini,

In response to comments from two referees, my coauthors and I revised our manuscript "The climate impact of ship NOx emissions: an improved estimate accounting for plume chemistry" (acp-2013-1033). Both referees provided helpful comments to improve and clarify our paper. Like our original submission, the revised paper addresses the important science question, "What are the climate impacts of human industries and transport methods?" We analyze the shipping sector in some detail, using global transport models and literature surveys. This paper therefore falls within the domain of Atmospheric Chemistry and Physics and should be of strong interest to this journal's readers once published. We appreciate the thoughtful comments of the referees and address each of them below in bold.

Sincerely,

Christopher Holmes

REFEREE 1

The authors presented and discussed a new representation to describe in a parametric form the dispersion of a plume emitted from a ship in the atmosphere. In chemistry- climate models, these processes needs to be treated with a sub-grid scheme. They claimed that the more accurate description including eight meteorological and chemical factors (end section 2.1) improves the calculation of the chemical species concentrations and the subsequent radiative forcing (RF). In my opinion, this research is interesting and timely since it attempts to show the role of small spatial and temporal scales in large scale processes. However, the research fells short in showing clearly and in an elaborate way the benefits of the new parameterization. Moreover the evaluation against observations is very short questioning the improvements in using the new parameterization. In consequence, the paper should largely improve to convince the reader that the new parameterization is necessary to be included in chemistry-climate models. Below, I include my main remarks.

The ship plume chemistry model and its implementation in the GEOS-Chem global CTM were described recently by Vinken et al. (2011). That earlier work did not calculate radiative forcing from ship NOx, which is the goal of this paper. A complete description of the model is therefore beyond the scope of this paper and already available in the literature. We do, however, describe the small updates to the model. Nevertheless, we have expanded our description of the model in the main text and supplementary material, as described further below.

1- As mentioned, the authors introduce a new parameterization, but the reader is left alone in the formulation and the sensitivity analysis.

a) Could they describe the equations/functions that forms the base of the new parameterizations and their dependences? Perhaps an Appendix is necessary to be included.

b) How sensitivity is the parameterization to the 8 factors mentioned? In my opinion, this is a key part of the research. The authors need to show which variables are relevant. Are the 8 factors equally important? Under which meteorological and chemical situations?

The model description by Vinken et al. (2011) already describes the parameterization in more detail than we could provide in an appendix. To aid the reader of this paper, we have added figures in the appendix showing how OPE, MOE and the fraction of NOx remaining depend on each of the 8 environmental variables. Vinken et al. (2011) already discuss the importance of the individual variables.

2- Clouds play a key role (stratocumulus, shallow and deep convection) in marine boundary layers and can regulate differently the dispersion and transformation of chemical species (Verzijbergh et al., 2009, Atmos. Chem. Phys. 9, 1289-1302). How are the dynamic and radiation effects of clouds included? In my opinion, a better description of clouds could be more beneficial that the new parameterization of dispersion. The authors need to discuss these aspects in their article.

The parameterization accounts for the effects of clouds on plume chemistry through photolysis rates. We include the following statement in Sect. 2.1.

"Clouds affect the parameterized plume chemistry through photolysis rates, but not through dispersion rates (Verzijlbergh et al., 2009)."

3- Closely connected to the previous point, and due to the lack of description of the parameterization. I am a bit surprise that an important sub-grid effect, the segregation of species, is not discussed neither included (Sykes et al., 1992, Atmospheric Environment 26A, 2565-2574; Galmarini et al., 1995, Atmospheric Environment 29, 87-95)? As far as I know, the limitation and inefficient mixing by turbulence can retard the chemical transformations in the first hours after emission. Could they explain if this process is included? If not, could they omit it?

Species segregation is not treated in the Gaussian plume model, but inefficient mixing by turbulence is limited to the first several minutes of plume aging. O_3 production and CH_4 oxidation occur mainly after this time period, so this model limitation has minimal impact on our results. We have added the following discussion of these issues in Sect. 2.1:

"Although Gaussian plume models poorly simulate the first several minutes of plume aging, when turbulent transport limits the rates of fast NO_x-O_3 chemical reactions (Galmarini et al., 1995; Sykes et al., 1992), they can provide a good representation of plume composition after about ten minutes (several kilometers) of aging, once turbulent dispersion homogenizes the plume (Galmarini et al., 1995). Indeed Vinken et al. (2011) demonstrated that their Gaussian plume model predicts NO_x, O_3, and OH concentrations consistent with field observations over several hours of ship plume aging (Chen et al., 2005)."

4- I miss throughout the paper a systematic validation (including uncertainties) with respect observations. I think it is fundamental to include this information to confirm the improvement of the new parameterization.

We address this in two ways. First, we show that the Gaussian plume model can reproduce the NO_x, O_3, and OH concentrations observed in the ITCT 2002 case study (Fig. S1). Second, we expanded the discussion of model vs. observations in Sect 2.1:

"The global CTM with updated plume chemistry has up to 3% less NO_x and 1% less O_3 in the marine boundary layer compared to the earlier parameterization. Therefore, comparisons of the CTM to observations over the North Atlantic and North Pacific Oceans

shown by Vinken et al. (2011; their Figs. 4,5) are unchanged. Specifically, in regions that are impacted by ship emissions but outside distinct plumes, the parametric plume chemistry predicts median NO_x abundances within 30% of observed values while instant dilution over predicts NO_x by a factor of 2. Ozone observations in the same regions are consistent with the plume parameterization but unable to falsify other model variants."

5- The last sentences of the conclusions are a bit confusing. The authors mentioned that there are uncertainties in the background atmosphere related to the emission and model formulation. What sort of uncertainties? Would it be better to explain these uncertainties (I guess related to clouds, non-uniform emissions,...) and place them if they are more important that the processes represented by the new parameterization?

We were referring to uncertainty in plume chemistry caused by the poorly known composition of environmental air that is entrained into the plumes, which we discussed at the end of Sect. 4. In addition, there are few detailed observational studies of plume chemical aging. We revised the last sentences of the conclusions to better explain our meaning:

"The largest contribution to this uncertainty arises from differing abundances of photochemical oxidants in the background atmosphere, which when entrained into ship plumes can alter their chemistry. Global emissions and model formulation both contribute to these differences in the background atmosphere. Further reductions in RF uncertainty are therefore unlikely without stronger observational constraints on radical sources and sinks in the remote marine atmosphere and additional observational case studies of ship plume aging."

REFEREE 2

General remarks: This study reveals the error of not including ship plume-chemistry in global chemical transport models and further the consequences on the estimation of radiative forcing from key greenhouse gases. The overall impression is that this is a solid and clearly presented study using sound and valid scientific methods giving credibility to the main conclusions. Uncertainties and challenges are to a large degree well explained though I think the need for better coverage of measurement data could be highlighted somewhat more. This still remains a challenge for a complete evaluation of different plume parameterizations and the error of just using instant dilution. I recommend publication but have some (mainly minor) comments I suggest the authors should address. See detailed comments below.

We agree that more extensive observations would enable better evaluation of plume chemical parameterizations. This is now stated in the conclusions: "The limited observations of ship plume composition during aging hamper efforts to widely evaluate the parameterization, but we have shown that it is consistent with available data."

Detailed comments:

Page 3429, Line 17: Figure 1 is cited here without being discussed. It is discussed much later in the manuscript. I suggest to remove the citation in the introduction and to change the figure order in the manuscript accordingly.

Done.

P3431, L4-5:"the most comprehensive and detailed global model". Though it could very well be correct this is a rather subjective claim as no objective comparisons with other models exist and there are a lot of issues going into NOx chemistry besides plume parameterization.

We deleted this phrase.

P3433, L8-9: Wind speed is added as a factor in the look up table. Since this is a new factor introduced in this study I miss some more information to the reader. How is it influencing CH4 oxidation and O3 production and how important is it compared to the other meteorological and chemical factors in the look up table (?)

We have added Fig. S2 in the Supplement to show how O_3 production and CH_4 oxidation respond to wind speed. In Sect. 2.1, we add "CH_4 oxidation and O_3 production can vary by a factor of 2 between wind speeds of 2 and 18 m s^{-1}."

P3434, L11: a= 0.34 is used. The origin and further use of the "a" factor on other models was a bit unclear. Is this the factor representative for GEOS-CHEM (i.e. model specific) based on an earlier calculation with a model version (Holmes et al. 2011) similar to the one used here? Is it this "a" factor that is used when the equation later is used to calculate the RFs from other models or is it the "a" factor representative for each model or an ensemble mean study (for the other parameters in the equation you use ensemble means from several from other studies). If you are using the same "GEOS-CHEM a factor" for all models how will this add to the uncertainty?

As is now explained in Sect. 2.2, "The *a* term derives from a literature survey of multiple CTMs and radiative transfer models (see SI and Holmes et al. 2011)" and is not specific to GEOS-Chem. We also added a section on "Radiative forcing from long-lived O_3 and CH_4" to the Supplement to provide more details.

P3434, L17-18: Why is the contribution from stratospheric water vapor not included? Though it is uncertain it is quite common to include in other studies quantifying methane associated RFs.

Stratospheric water vapor is now included in all of our estimates of CH_4 RF. Text, tables and figures are all updated accordingly.

P3435, L15-16: Why is not surface deposition of NOx included in the Gaussian plume model? Wind speed was added as an extra factor in the look up table and it should not be so much effort to include deposition. I suggest including it in future studies as not having it certainly adds some unnecessary uncertainty to the calculations.

We tested the effects of adding dry deposition of NOx to the Gaussian plume model. It turns out that N_2O_5 is already rapidly consumed on sulfate and other aerosol in the ship exhaust, so neglecting dry deposition had negligible impact on our results. Other NO_x species deposit too slowly to affect the plume chemistry over 5 hours.

P3440, L1-4: The discussion of a cancellation in the RF of sulfate and nitrate components is a bit misleading. Indirect effects have been shown to be very important for shipping in previous studies. It should be stated more clearly that you haven't included indirect effects and that these might be important.

At the end of this paragraph, we added, "Aerosol indirect effects, black carbon, and organic carbon also contribute to radiative forcing from ships (Eyring et al., 2010) but are beyond the scope of this study of ship NO_x."

P3440, L6: "our RF estimate. . .. . .to be the most realistic RF estimate to date...". Again a subjective statement. See earlier comment.

The revised sentence says, "Our global RF calculation using parametric plume chemistry is the first to account for sub-grid scale ship NO_x chemistry."