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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The climate impact of ship NO_x emissions: an improved estimate accounting for plume chemistry" *by* C. D. Holmes et al.

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Both referees have provided helpful comments to improve and clarify our manuscript. We appreciate their thoughtful comments and address each of them below in bold.

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. O3 production and CH4 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 NOx-O3 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 NOx, O3, 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 NOx, O3, 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 NOx and 1% less O3 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 NOx abundances within 30% of observed values while instant dilution over predicts NOx 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."

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 3427, 2014.