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Comment

Interactive comment on “Accounting for non-linear chemistry of ship plumes in the GEOS-Chem global chemistry transport model” by G. C. M. Vinken et al.

Anonymous Referee #1

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The presented study deals with accounting for the non-linear chemical processes during the dilution of plumes emitted from ships in a global CTM. It uses a plume-in-grid formulation to calculate the species concentrations in the plume after it dilutes to the model's scale under different meteorological and chemical conditions. The results are used to build a look-up table that serves to provide modified NO_x emissions and emissions of secondary species HNO₃ and O₃, replacing the instant injection of the original emissions, a common practice in most global and regional CTMs. The performance of the method is validated against aircraft based marine boundary layer measurements. The topic of the study definitely fits the scope of ACP and lines up with previous papers in ACP dealing with ship emissions environmental impact. Therefore I recommend the

C6168

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Discussion Paper



publication in ACP. However, there are several issues below that have to be taken into account before.

General comments

1) It is correct that the paper offers a comprehensive description of the method how the chemical non-linearities in the diluting plume are taken into account, but in my opinion very few space is given to the discussion of its limitation. The "fraction of NO_x remaining" and integrated NOPE are in fact a function of the environmental conditions neither at the initial release time, nor at the actual emission into CTM (5h later), but of their entire evolution during the dilution. Although authors assume that these parameters do not change "dramatically" during the plume dilution, the reality can be very different (e.g. photolysis rates can drop significantly when moving below a cloudy area during the first 5 hours after the injection). How would the model results be affected - improved, if some integrated (averaged) environmental parameters were accounted for, i.e. the global CTM "remembered" the environmental conditions from the last 5 hours of integrations?

2) Although the majority of the abstract concerns the results gained by the new method implemented in GEOS-Chem, the presentation of the actual simulations occupy only two pages in the manuscript. I would suggest to extend this part of the paper by a couple of additional experiments/results: e.g. why the standard model with ship emissions (that replaces 1 NO_x molecule released by 1 HNO₃ and 10 O₃ molecules) was not applied here for comparison of the results (like in the evaluation)? Further, as instant dilution leads to overestimated ozone production and NO_x lifetime, this consequently increases OH concentrations, leading to overestimated CH₄ lifetime decrease. It would be thus interesting to see, how OH is perturbed by the parametrization (and eventually by the introduction of ship emissions themselves, with respect to the non-ship case). What is the vertical extent of the ship NO_x impact as well as the impact of the parameterization itself? - a zonal plot or a longitude cross-section across the Atlantic would be interesting to add. Further I think the results have to

C6169

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be discussed in more details, putting the findings into context using the references from the "Introduction" and eventually others. The "Conclusions" further should in my opinion discuss also the future potential improvements of the method considering its limitations (see General comment 1)

Specific comments

P17792, L5-8: The reference on Charlton-Perez et al. work is correct but the context why this sentence is included might be not clear. Is it because of the expectation that by increasing resolution, models start to resolve plume processes so, in accordance with the ship plume studies, produce less ozone?

P17792, L14: Why the authors start the paragraph with "Not a single global CTM currently takes the in-plume effects during ship plume dispersion into account", if just a few lines below they refer to GEOS-Chem model in early stages and GMI which actually already account for these processes by not emitting directly NO_x, but O₃ and HNO₃?

P17793, L9: Huszar et al. (2010) did not apply the method of effective emission indices but the approach of effective reaction rates (Paoli et al., 2011).

P17798, L20: Sensitivity analysis: the section already starts with the sensitivity analysis of certain parameters on "fraction of NO_x remaining" and integrated NOPE, but the reader is left with questions what implications led the authors to choose these parameters and not others. Authors state at P17797, L8 that the sensitivity analysis is performed in order to determine the most critical parameters, but in fact the parameters are already determined before the sensitivity analysis, at least this impression comes to reader.

P17799, L13-14: Why the authors do not simply examine the dependency on J(O₁D), as sensitivity test on J(NO₂) is already included?

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Interactive
Comment

P17799, L15-23: Firstly, I am not sure by the importance of using solar zenith angle θ_0 as parameter to include in the look-up table. Isn't the photolysis rates $J(\text{O1D})$ and $J(\text{NO}_2)$ already contain the relevant information. e.g. at night they equal to zero, at noon they are at maximum (considering clear sky conditions)? Secondly, is the reason to include θ_5 as environmental parameter because we cannot assume about solar zenith angle not to change "dramatically" (P17802, L26)?

P17800, L16-21: What are the intervals for the LUT parameters and for how many values from the particular interval was PARANOX run? Was the LUT constructed for every possible combination of these parameters (which could lead to very large number of PARANOX runs; e.g. 10 possible values for each parameter - 10^7 runs, assuming independency between them which of course is not completely true.) Further I assume that environmental parameter (e.g. temperature, background NO_x , O_3 etc.) were constant in time during a PARANOX run. However, it is not clear, how the solar zenith angle evolved between the examined values θ_0 and θ_5 . Were there some real evolutions corresponding to different release hours and latitudes, as authors state later in Figure 2. caption?

P17800, L22 - P17801, L8: Authors claim that frequency of situations with weak winds, low marine boundary layer, strong emissions leading to saturation effects is small, hence they did not include these parameters in the LUT. However, what about strong winds and strong vertical mixing, how can this influence the plume dilution into the background air, consequently the evolution of chemical species? And what about dry/wet deposition of plume species? As stated at P17794: L14, PARANOX does not consider rainout, does it hold for dry deposition as well? What is the expected effect of these simplifications? Further, why is water vapour not included in the sensitivity analysis? It can have impact on reactions (R1) and (R12) influencing OH radical formation and NO_x nighttime chemistry (HNO_3 formation).

P17802, L6: The depth of the lowermost layer should be specified first here (it is done later at P17805, L11). How can this affect the results? If the lowermost layer is too thin,

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at certain conditions (higher mixing depths) the ship plume will dilute into higher layer(s) as well within 5 h after release. Emissions in GEOS-Chem were injected always in the lowermost layer? What is vertical model extent (the altitude of the 47th layer?).

P17802, L14-22: Authors state that no daily variation of emissions was considered, only monthly variation in case of EMEP emissions. What about hourly variation, was it considered? if yes, than the original emissions that are reduced have to be those from 5 h before actual model time. This raises also the question of the emissions input frequency.

P17802, L8: Was the spin-up run (2004) common for each experiment or they were all run with spinup, but only the second year of each run (2005) was analyzed? Did this year (2005) represent an average climate or did it encounter some extremities compared to other years?

P17804, L1-9: Additional simulations: the "standard model" is used with no-ship emissions, but why it is not used (as it already suggested in the general comments) with ship emissions as well, to compare it with the new approach and with the instant diluting case (so having 4 simulations: 1 without ship emissions, 3 treating ship emissions in a different way)? Further it is difficult to follow what simulations were performed. It would be helpful to assign ID for each run in a similar way as it was done in e.g. Cariolle et al. (2009) or Huszar et al. (2010). The presented absolute and relative changes than can be explicitly expressed by these IDs.

P17805,L8 - P17806, L18: I would welcome a more systematic comparison. Authors are presenting first the effect of parameterization on NO_x in absolute and relative sense for January and July. Why there are no "relative" figures for ozone (only absolute ones)? It would be useful to gain impression of the relative importance of ozone change caused by the parameterization. Further, the effect of ship emissions are presented, but only for summer. Why is winter omitted? At last, in my opinion a more common approach would be to present first the ship emission induced NO_x and O₃ changes (using either

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the improved model or the instant diluting one) and than the parameterization's effect, to compare the magnitude of the two (i.e. how the ship induced NO_x/O₃ changes are modulated by the parameterization). However, I understand that presenting all these (absolute/relative) differences, including the additional standard model run (see previous comment), eventually vertical crosssections, would significantly increase the number of figures. So I at least suggest to show the relative difference of the ozone change due to parameterization and the effect of ship emissions for winter.

P17816, Figure 2 and P17817, Figure 3: in the 6th and 7th panel: solar zenith angles 0 - 90 deg correspond to darkness? In text (e.g.P17799, L16) $\theta_0 = 10$ deg is attributed to noon at low altitudes so this is clearly a mistake and the x-axis have to be relabeled.

Technical corrections:

P17796, L13-14: Starting at this point the “fraction of NO_x remaining” is used trough the whole study. I suggest to use “remaining fraction of NO_x” or “plume-fraction of NO_x”.

P17790, L7: Here and further in the manuscript the “in the 5h after the release...” formulation is used. I suggest to use rather “during the first 5 h after plume release” (like it is used in von Glasgow et al., 2003)

P17799, L22: the name of the parameter (θ_5) missing

Figures: I think the spatial figures 6., 7., 8. and 9. can be reduced in size a bit in order that the two sub-figures fit next to each other without losing the resolution unacceptably. With this, more figures can be showed (as I proposed earlier) without increasing the number of pages significantly.

References:

Cariolle, D., Caro, D., Paoli, R., Hauglustaine, D. A., Cuenot, B., Cozic, A., and Paugam, R.: parameterization of plume chemistry into large-scale atmospheric models: Application to aircraft NO_x emissions, J. Geophys. Res., 114, D19302,

doi:10.1029/2009JD011873, 2009.

Paoli, R., Cariolle, D., and Sausen, R.: Modeling and computation of effective emissions: a position paper, *Geosci. Model Dev. Discuss.*, 4, 137-196, doi:10.5194/gmdd-4-137-2011, 2011.

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