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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Source identification and budget analysis on elevated levels of formaldehyde within ship plumes: a photochemical/dynamic model analysis" *by* C. H. Song et al.

**Anonymous Referee #2**

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**General comments:**

This paper uses a photochemical/dynamical model to analyze the budget of elevated levels of formaldehyde within ship plumes. The model has been developed, described and evaluated previously by Kim et al., ACP, 2009. As argued by Kim et al., the difference to previous boxmodel studies (e.g. Chen et al., JGR, 2005; Song et al., 2003; von Glasow et al., 2003) is that the photochemical/dynamic model



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actually accounts for the photochemical aging of the ship plume while considering the variations in the concentrations of background species. Kim et al. focused on the model description and an evaluation of the model results with respect to NOx, NOy, ozone, HNO3, and H2SO4. The study here focuses on the source identification of formaldehyde. Since discrepancies between global chemistry model simulations and observations have been found for ship-induced tropospheric column enhancements of HCHO (Marbach et al., ACP, 2009), and since these discrepancies have not yet been resolved or fully understood, this is an important process study which fits well within the scope of ACP.

The paper is generally well structured, but could be improved in terms of clarity, for example (1) some of the conclusions seem to be generalized, (2) some of the figures are not very well described, and (3) the text can be shortened and be written more precise in some sections (see detailed comments below). Kim et al., which is the companion study of this paper, claim that the presented ship plume chemistry model should be used to model the changes in ship-plume chemical compositions and to better evaluate the atmospheric impact of ocean-going ship emissions. While the model adequately simulates some aspects, a caveat should be mentioned, that the presented model simulations will always be limited to specific case studies for certain regions; one weakness of the case study simulations is the background which is often not very well known and initialized from a variety of different sources (CTMs, observations, etc.); this could cause inconsistencies or the neglect of seasonal variations in the background as done here. Therefore a full assessment of the global impact of ship emissions will in addition require global model studies and observations. The paper could therefore do more in establishing ties between global models and the model studies presented here. Overall, I can recommend publication of the paper in ACP once my comments below have been adequately addressed.



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**Specific comments:**

Abstract: the one main conclusions of the manuscript is that CH4 oxidation by elevated levels of in-plume OH radicals was found to be the main factor responsible for the elevated levels of HCHO in the ship corridors. The authors should clarify that this conclusion refers to (it seems) a single ship under a certain meteorological condition. Similarly, the second main conclusion of enhanced HCHO levels in different regions seems to be based on case studies carried out under 10:30 am local time. While this is fine, the abstract (and summary) should avoid generalization of these statements that are not justified by the manuscript.

p. 15444, l. 14-25: This paragraph does not appropriately summarize the main conclusions of Marbach et al. Rather Marbach et al. already point to several of the key conclusions that are found here, so this study in many ways confirms some of the Marbach et al conclusions or indications. This should be made clearer. For example, Marbach et al find differences between the observations of NO2 and HCHO and conclude that these differences have important implications for the source of the enhanced HCHO concentrations. From the differences between the simultaneously observed tropospheric column NO2 values over the shipping route to those of HCHO they conclude that direct emissions of HCHO (source 1 of elevated HCHO in this study) or degradation of emitted NMHC (source 2 of elevated HCHO in this study) cannot explain the observed enhanced HCHO values and point to increased CH4 degradation due to enhanced OH concentrations related to the ship emissions (source 3 of elevated HCHO in this study).

Again on the same point, Marbach et al. conclude that 'the CH4 oxidation source is probably too weak to fully explain the observed values'. The authors should clarify their results in light of this statement. Do they disagree with it also in the context of global model simulations? If yes, why? Since the authors also exclude source 1 and



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2, could other factors as speculated by Marbach et al. play a role? Then this should be brought forward to the abstract.

As mentioned above, the paper could do more in comparing the results to previous global model simulations and conclusions, which would also make the paper more useful in a broader context. How would the results change if the emissions were instantaneously emitted on to a large grid box? Is it possible to assess this with the model presented here? If so this would be a valuable extension which could at least be mentioned in the outlook if it is beyond the scope for this study.

p. 15445, l. 14-21: rather than explaining the different options this model has, some more details (even if they repeat Kim et al.) on the model as used in the setup here would be helpful. The paragraph as it stands is not sufficient to understand the model simulations from this paper. Please extend. I would recommend to introduce a separate subsection '2.1 Model description' and rename the current headline with e.g. '2. Model characteristics, evaluation and simulations'.

p. 15445, l. 23 - p. 15445, l. 12: This paragraph should be moved from '2.2 Model validation' to '2.1 Model description'.

Figure 2: The figure caption says that the NOx emission rates have been varied from 2.6-13.3gs-1, but it is not clear in the figure which lines or shaded areas correspond to this range. Also there are quite some discrepancies between observed and modeled values in particular for HNO3 which should be discussed.

p. 15448, l. 10: I would recommend dividing the section on 'Model simulation' into two subsections, '2.2.1' on the base-case (as it stands now) and '2.2.2 Constructed model simulations' (i.e., move the model description from Section 3.2 but not the results to Section 2.2.2). In this way all model simulations are introduced in



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Section 2.2. which gives the reader more guidance to understand the results that are described in Section 3.

p. 15447, l. 11 - p. 15448, l. 5: How do these values compare to the most up-to-date study on ship emissions by the International Maritime Organization (IMO)? The Second IMO GHG study provided an update of emission factors and total emissions separated into various ship types and species:
Buhaug, Ø., J. J. Corbett, Ø. Endresen, V. Eyring, J. Faber, S. Hanayama, D. S. Lee, D. Lee, H. Lindstad, A.Z. Markowska, A. Mjelde, D. Nelissen, J. Nilsen, C. Pålsson, J. J. Winebrake, W.–Q. Wu, and K. Yoshida, Second IMO GHG study 2009; International Maritime Organization (IMO) London, UK, March 2009, see http://www.imo.org/Environment/
The assumptions here should be put into context of these results, e.g. the NMVOC emission factor was given as 2.4 kg of NMVOC per tonne of fuel and NOx for Slow-speed diesel engines 90-85 and for Medium-speed diesel engines 60 to 51 kg per tonne of fuel. The IMO study therefore indicates that the NOx:NMVOC ratio varies for different ship types which should be considered for the constructed cases. It should also be made clear in summary and abstract that the base-case study is valid only for a certain ship.

p. 15450, l. 10-12: this seems incorrect.

p. 15450, l. 1-28: a lot of this is repetition of issues that have already been discussed in the introduction, so should be removed here. Rather this section would be more readable if it started with the actual results before they are discussed (i.e. directly start this section with p. 15451, l. 5).

p. 15451, l. 7-10: what about non-linearities between these four cases?



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Figure 3: It is hard to understand this figure with the limited explanation that is given. Please expand and maybe draw the readers' attention specifically to some of the lines shown in the plot (e.g. compare line X with line Y at the end of the sentence in line 13). Maybe this figure should also be better split into two figures that show the first two rows and the second two rows in separate figures.

p. 15453, l. 9-29: as for Figure 3, the description of this figure has to be improved to give the reader a bit more guidance in understanding the results. Some of the panels are really small (I can hardly see any differences in Figure 4a between the cases).

Figure 5: now the figure caption refers to (a) OH etc whereas in Figure 4 (a), (b) etc was referring to the individual rows. A similar format would help; also write in figure caption OH red line, methane lifetime blue and HCHO production rate green line (same in Fig. 4).

p. 15454, l. 5: while equations 1 and 2 are clear from the reactions given in Table 2, this is not clear for equation 3. Provide details that confirm this equation is correct; the given text is rather confusing.

p. 15458, l. 17-21: it seems that the background conditions are chosen to be the same for all seasons. This is obviously a significant weakness and caveats should be mentioned.

Table 4: remove digits after the comma since this gives the impression that the simulations actually have this accuracy.

p. 15460, l. 10-13: Marbach et al noted significant difference in the tropical regions due to the fact that the ship track corridor is traversed by the Intertropical



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Convergence Zone (ITCZ) twice a year. As a result, the mean wind directions of summer and winter are nearly opposite with corresponding effects on HCHO. Add a comment.

p. 15463, l. 1: the first sentence sounds like this is a result from this paper. Make clear that this is a result from previous studies.

p. 15463, l.1-16: The majority of the results are derived from the base case which is examined for a certain ship and a certain meteorological situation. Additional sensitivity simulations are carried out for 10:30am local time. It should be made clearer in the summary and abstract which results are derived from the base case and which for the extended set. All figures are shown only for the base case, and the detailed analysis that is presented under Section 3.1 is not repeated for all sensitivity scenarios; rather for those only the HCHO enhancement is studied. This needs to be much better summarized and a generalization of statements avoided.

p. 15463, l.17 - p. 15464, l. 11: This is all rather speculative and shouldn't make up half of the summary.

**Minor comments:**

p. 15442, l. 4: 'predicted by global 3-D chemistry-transport models' should be replaced with 'simulated by global 3-D chemistry-transport models (similar replace or remove predicted in the context of model simulations throughout the manuscript).

p. 15443, l. 20: 'vertical columns' should be replaced with 'tropospheric columns'.

p. 15443, l. 24: The sentence 'Moreover, if the main. . .' should be rewritten.



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p. 15444, l. 6-8: The Corbett and Koehler, 2003 reference is on ship emissions, while in the context of this sentence it reads like this paper is on CH4 destruction in the MBL.

p. 15444, l. 9: TBL?

p. 15444, l. 9-13: The paragraph on the Hoor et al paper is confusing. Global models consider non-linearities and chemistry-climate interactions, but so far do not include a parameterization or modeling of subgrid scale processes in ship plumes. This should be clarified.

p. 15444, l. 20: 'Considering. . ..': for readability, start new paragraph.

p. 15460, l. 6: 'observed' should be 'simulated'

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 15441, 2010.