

Author's comments to anonymous referee #1

First of all, thank you for your valuable comments and suggestions. In the revised manuscript, we attempt to improve the manuscript based on your comments and suggestions. Major changes made in the manuscript are as follows:

- (1) We re-drew Figs. 2 through 6.
- (2) We added Fig. 8.
- (3) We split original Fig. 3 into Figs. 3 and 4, in order to more clearly show two separate issues: the changes in the ship-plume HCHO concentrations (Fig. 3) and the source budget of the ship-plume HCHO concentrations (Fig. 4).
- (4) We removed the simulation results from neutral MBL condition for the ITCT 2K2 ship-plume case, in order to more consistently show the results from Fig. 3 to Fig. 9. In Fig. 6, we added the simulation results from stable MBL condition.
- (5) We decided to show the results for only six ship-plume transects in Figs. 5 and 6, in order to more succinctly present our simulation results.
- (6) We inserted color-codes into Fig. 6.
- (7) We re-constructed Sect. 2.3. Now, Sect. 2.3 has three subsections: (1) Estimation of the emission rates; (2) Model simulation for base case; and (3) Model simulation for constructed cases.
- (8) We re-constructed Table 3.
- (9) We shrank down many parts of the text.

Other added/modified parts in the manuscript are painted in a red color in the revised manuscript. Here, we would like to reply to some specific questions raised by you below:

1. *“Based on the levels given in Table 3, the rate of NMVOC reaction with OH would be a significant fraction of the CH_4 oxidation rate. The reaction rate coefficients are 3 – 4 orders of magnitudes higher for the NMVOC reactions than that for CH_4 . The CH_2O yield (mostly through Acetaldehyde) can be significantly higher than unity. Thus, it is not easy to understand why the difference between Case I and II is not observable in Figures 3 and 4.”*
- The concentrations in Table 2 are the background concentrations used in this study. If one calculates the budget of HCHO with these concentrations, for example, using a steady-state model, about 10-20% of the HCHO concentrations come from the NMVOC oxidations. We added an analysis in the revised manuscript that at least ~12% of HCHO is produced from the NMVOC oxidations for the ITCT 2K2 ship plume case (refer to newly added Fig. 8). However, this contribution appears to be from the background NMVOC concentrations. NMVOCs emitted from the ship do not greatly affect the enhancements of the levels of in-plume HCHO, since NMVOCs are composed of many different individual NMVOC species

and thus are rapidly diluted. Moreover, the NMVOC species are diluted during the OH depletion period. These results are against what one usually expects. This was why we put a rather lengthy explanation into the manuscript regarding this issue (please, refer to pp.14:18- pp.15:9; pp.19:20-pp.20:5).

2. *“In addition, freshly emitted CH₂O can be a net source of HO_x. It is desirable for the authors to show the OH values for all three cases.”*

- Yes, we showed the OH mixing ratios in Fig. 6.

3. *“As for the NMVOC contribution, the authors should conduct a detailed budget analysis for CH₂O, including a budget for CH₃O₂ and CH₃OOH to show the relative contributions from NMVOC and CH₄ and direct CH₂O formation throughout the NMVOC degradation steps.”*

- We carried out a budget analysis. Please, check out Fig. 8 and refer to pp.19:20-pp.20:5.

4. *“Based on the background mid-latitude conditions given in Table 3, a quick box-model calculation gives NMVOC contribution in the order of 10%. The equation (3) cited in the manuscript is neither rigorous nor practical (see details in specific comments). The term Φ_i is rather difficult to determine as it is a function of chemical conditions.”*

- Obviously, Eq. (3) is not practical. Rather than, it is a conceptual expression for estimating the HCHO formation rate. Actual equation we used in this study was the following one from the modified Lurmann condensed chemical mechanism:

$$\begin{aligned}
 F_{\text{HCHO}} = & k_{\text{CH}_3\text{O}_2+\text{CH}_3\text{O}_2}[\text{CH}_3\text{O}_2]^2 + k_{\text{CH}_3\text{OOH}+\text{OH}}[\text{CH}_3\text{OOH}][\text{OH}] + k_{\text{CH}_3\text{O}+\text{O}_2}[\text{CH}_3\text{O}][\text{O}_2] \\
 & + k_{\text{CH}_2\text{OH}+\text{O}_2}[\text{CH}_2\text{OH}][\text{O}_2] + k_{\text{RAN1}+\text{NO}}[\text{RAN1}][\text{NO}] + 0.5k_{\text{ISOP}+\text{O}_3}[\text{ISOP}][\text{O}_3] \\
 & + 0.9k_{\text{RIO2}+\text{NO}}[\text{RIO2}][\text{NO}] + k_{\text{INO2}+\text{NO}}[\text{INO}_2][\text{NO}] + 0.5k_{\text{MVK}+\text{O}_3}[\text{MVK}][\text{O}_3] \\
 & + 0.3k_{\text{VRO2}+\text{NO}}[\text{VRO2}][\text{NO}] + k_{\text{MVN2}+\text{NO}}[\text{MVN2}][\text{NO}] + 0.65k_{\text{MVCR}+\text{O}_3}[\text{MVCR}][\text{O}_3] \\
 & + k_{\text{MAN2}+\text{NO}}[\text{MAN2}][\text{NO}] + k_{\text{ETHE}+\text{O}_3}[\text{ETHE}][\text{O}_3] + 2k_{\text{EO2}+\text{NO}}[\text{EO2}][\text{NO}] \\
 & + 2.4k_{\text{EO2}+\text{EO2}}[\text{EO2}]^2 + 0.525k_{\text{ALKE}+\text{O}_3}[\text{ALKE}][\text{O}_3] + k_{\text{PO2}+\text{NO}}[\text{PO2}][\text{NO}] \\
 & + k_{\text{PRN1}+\text{NO}}[\text{PRN1}][\text{NO}] + k_{\text{CHO2}+\text{NO}}[\text{CHO}_2][\text{NO}] + k_{\text{CHO2}+\text{NO}_2}[\text{CHO}_2][\text{NO}_2] \\
 & + k_{\text{CHO2}+\text{SO}_2}[\text{CHO}_2][\text{SO}_2] + k_{\text{MCO3}+\text{CH}_3\text{O}_2}[\text{MCO}_3][\text{CH}_3\text{O}_2] + k_{\text{MCO3}+\text{CH}_3\text{O}_2}[\text{MCO}_3][\text{CH}_3\text{O}_2] \\
 & + J_{\text{MCP}}[\text{MCP}] + 0.5k_{\text{MCP}+\text{OH}}[\text{MCP}][\text{OH}] + k_{\text{EP}+\text{OH}}[\text{EP}][\text{OH}] + 2J_{\text{EP}}[\text{EP}] \\
 & + k_{\text{HACO}+\text{NO}}[\text{HACO}][\text{NO}] + k_{\text{HEP}+\text{OH}}[\text{HEP}][\text{OH}] + 2J_{\text{HEP}}[\text{HEP}]
 \end{aligned}$$

Although this is the true expression we used in this study, we felt that showing the entire equation in the manuscript appears to be clumsy. That was why we used a rather conceptual expression of Eq. (3). Now, Eq. (3) is modified. Please, check out Eqs. (3), (3-1) and (3-2). Also, see pp.17:14-18:3.

5. *“Finally, the authors should also consider the background Acetone and Ketone in the analysis, since oxidation of these species can be a non-negligible source of CH₂O.”*
 - Both may also be non-negligible sources of HCHO. But, we did not take these two species into account in this study. There were a couple of reasons. In the ship-plume modeling, the two species are got into the ship-plume volumes by entrainment process from the background air. However, the concentrations of the two species were not measured during the ITCT2K2 ship-plume experiment campaign, so that we could not constrain our ship-plume model. In addition, since both species are reservoir species, chemistry may not be very active over the ship-plume transport time-scale of “140 min”.

Specific comments

1. *“Pg 4, line 22: “the removal of CH₄ can also ...”, is this an over-simplified statement?”*
 - We re-wrote the sentence. Please, see pp. 4:20-pp.5:2.
2. *“Pg 5, paragraph 2: This paragraph really casts doubts on if there is a CH₂O enhancement in the ship traffic corridor. The authors should re-organize this paragraph and clarify their points.”*
 - We re-wrote the sentence. Please, see pp. 5:3-5:13.
3. *“Pg 7, line 16/17: OPE is not a direct observation”*
 - We re-wrote the sentence. Please, check out pp. 8:11.
4. *“Pg 8, paragraph 1: The authors should provide more detailed discussions on the comparison between model and observations, shown in Figure 2. It should be pointed out that the level of agreement is different for different species and ship plume transects.”*
 - We put more discussions into Sect. 2.2, but many discussions had already been made in the previous publication (Kim et al., 2009). Please, check out pp.9:4-pp.9:10. Also, as mentioned above, we re-drew Fig. 2.
5. *“Pg 11, line 8: “O(¹D) radicals mainly react with the more” This is not correct, the main losses for O(¹D) are the reactions with O₂ and N₂.”*
 - We revised the sentence. The primary reactions of O(¹D) are quenching reactions with N₂ and O₂. Please, check out pp.13:16-13:18.
6. *“Pg 16, equation (3): This equation is incorrect since there are important feedback reactions after CH₃O₂ + HO₂ and CH₃O₂ + CH₃O₂, which will produce CH₂O. The authors should*

have some discussion on the values of the CH₂O yield from the key NMVOC species from ship emissions. This equation should be revised or deleted. A better definition of NMVOC contributions is need for the manuscript. The authors should trace sources of CH₃O₂ and CH₃OOH in terms of relative contribution from NMVOC and CH₄ oxidations.”

- As mentioned in the previous reply, we modified Eq. (3). As also mentioned above, we modified Table 3. In new Table 3, you can find the feedback reactions from CH₃O₂ + HO₂ and CH₃O₂ + CH₃O₂. Please, check out pp. 17:4-pp.18:22.
7. *“Pg 37, Table 3: The CO levels appear to be very high for the tropical case! The authors are referred to check NASA PEM-Tropics A and B observations. The other CO values are more consistent with the polluted regions. This reviewer questions if the authors should show the ship emission impact in polluted environments?”*
- No, we did not (cannot) consider the impacts of other ship emissions on the ship-plume photochemistry. The elevated levels of CO are possibly due to biomass and bio-fuel burnings, both of which are active in South Asia.