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Interactive comment on “Pollution plumes observed during CARIBIC flights in the upper troposphere between South China and the Philippines” by S. C. Lai et al.

Anonymous Referee #1

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General comments

The CARIBIC program is a unique and important measurement program in the upper troposphere – lower stratosphere (UTLS) region providing a rich and growing database of relevant trace gases and aerosols. This paper describes measurement results for a suit of trace gases and aerosols from a number of flights over Southeast Asia where the aircraft encountered several pollution plumes in the upper troposphere. The data presented here are a relevant contribution to the relatively scarce body of in-situ data available for the troposphere over Southeast Asia. The discussion mainly focuses on enhanced levels of CO, CO₂, CH₄, non-methane hydrocarbons (NMHCs)

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and aerosols. Finally, the authors make an estimate of the anthropogenic contribution to these plumes.

In general the paper is clearly written and the level of English is adequate. However, on several points the discussion on the results is or unclear is too concise and needs further explanation to improve the clarity and comprehensiveness of the paper. More specifically, the authors ignore the important role of biofuel emissions in Southeast Asia and mainly write about “biomass burning emissions” in general terms. The authors should elaborate more on the context of their observations in the light of earlier studies (TRACE-P, PEM West B). Also the role of these plumes for the composition and chemistry of the upper troposphere over Southeast Asia (focusing on the role of ozone and aerosols) should be addressed in more detail in the light of results from earlier studies.

Hence, I recommend publication of the paper in ACP following the modifications as suggested below.

The paper needs revision focusing on the following items:

1. The paper mainly talks about “biomass burning” versus “anthropogenic emissions” generally ignoring the distinction between “natural” biomass burning (e.g. forest fires) and “anthropogenic” biofuel emissions which are a very important source in Southeast Asia (e.g. papers by Streets et al., 2003). MODIS fire maps only indicate that there were fire hot-spots which could be either natural or man-made (e.g. deforestation activities). It doesn’t tell you the real extend of the biomass burning emissions in Southeast Asia not visible on these fire maps. Here you have to refer to studies by Streets et al. and use the RAINS-Asia emission data base.
2. What is the context of the result? The data could be compared more comprehensively to results from previous studies conducted over the same area. Elaborate

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- more on how the CARIBIC results compare to other observations from earlier studies conducted over South-East Asia (TRACE-P, PEM West B). How representative are your results of Asian emissions? See also point 5).
3. How important are these plumes for the composition of the UT/(-LS) region e.g. with respect to the ozone and aerosol budget (both important for the radiative budget)? The authors should discuss in more detail the $\Delta\text{O}_3/\Delta\text{CO}$ ratios in the light of earlier studies (Jost et al., 2003; Kondo et al., 2004).
 4. Include an additional graph with vertical profiles for the 4 different flights for CO, O₃, aerosols, CH₃CN and acetone to elucidate the vertical structure of the plumes.
 5. Include an additional table where you present the mean concentrations of the relevant gases and aerosols for the plumes ((W15, W17, W19 and W21) as well as the background condition (W18) and compare these to data from Blake et al. (rather than including W18 in table 1).

Specific comments

Page 4, line 9-10: “It is expected that in addition to anthropogenic emissions, oceanic and biomass burning emissions also influence atmospheric composition in this region.” The authors should refer to the fact that in several earlier studies the role of oceanic and notably biomass burning emissions has been pointed out (e.g. Woo et al., JGR, 2003; Kondo et al., JGR, 2004). Write: “Based on earlier studies (e.g., *refer here*¹) it can be expected that . . . composition in this region.”

¹Suggestions for references on oceanic emissions:

C. Warneke, and J. A. de Gouw, Organic trace gas composition of the marine boundary layer over the northwest Indian Ocean in April 2000, *Atmospheric Environment* 35 (2001) 5923–5933

Page 4: last line of the Introduction: “The contribution of regional sources to these plumes is estimated here.” Here the authors should also mention the other topics as suggested in the Specific comments above (relevance, impact, context of the results).

Fig. 2a and 2b: The graphs are too small and should be enlarged for better reading. Please include the appropriate air sample names (W14 to W21) in the graph for clarification.

Page 5, line 14: Include “different” before “laboratories”.

Page 7, line 1 and 2: For CH₃CN and acetone it is written “not shown in Fig.2”. They are however present in Fig.2a and not indeed not in Fig. 2b. Please correct this.

Page 7, line 17: Change “strong plume” to just “plume”. The classification strong is not so relevant here.

Page 7, line 22-23 to page 8, line 1-3: “A strong decrease in trace levels. . . .than in sample W15”. The explanation is confusing and should be clarified. Which trace levels do are meant here? By looking at the graph it appears that the aircraft reaches cruise altitude while leaving plume 2. Trace gas levels are increasing when entering plume 3 at cruise altitude while ozone decreases.

Page 8, line 11-17: See also point 5) of the Specific comments above. Here the authors mention sample W18 containing “close to the background levels reported during previous campaigns”. Since the authors refer to the background levels reported by Blake et al. (1997) I suggest they include a separate table were you present the concentrations of all the relevant trace gases from W14 together with the background data from Blake et al. for comparison. In the same table the authors can present the mean and standard deviation of the relevant trace gases from the plume air samples (W15,

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W17, W19 and W21) and compare them as well to results from Blake et al.

Page 9, line 13-14: Use round numbers for the acetonitrile and acetone values (2597 instead of 2596.9). This level of accuracy is not relevant here and is at the same time questionable for the PTRMS technique.

Page 9, line 16-19: “To summarize,. . . .relative to the other samples (W14, W16, W18 and W20).” Why are ozone and aerosols not mentioned here. Why is that? Please include.

Page 9, line 22: “As in other studies, . . .”. What other studies? Include some references here.

Page 10, line 5-7: “When air is in contract. . . ., would be expected.” Include a reference here.

Page 10, line 10-17. I find the evaluation of the $\delta^{13}\text{C}$ results written in an disorderly and unclear manner. Please explain the meaning of $\delta^{13}\text{C}(\text{CO}_2)$ range you find (-8.52% to -8.31%) and include a reference. Change “High CO_2 values always correspond to low $\delta^{13}\text{C}(\text{CO}_2)$” to “Elevated CO_2 concentrations generally corresponds to low $\delta^{13}\text{C}(\text{CO}_2)$” and try to include a reference.

Page 10, line 17: Include “the NMHCs” before “ C_2H_6 , C_3H_8 ,. . .”

Page 10, line 19: Explain were the NHMCs are usually increased.

Page 10, line 22-23: Clarify what “a slight increase in O_3 ” means by including the actual increase relative to the “background”.

Page 11, line 1-2: Rephrase this sentence it is unclear like it is. When you include a table (as suggested in the Specific comments) with actual values for the different aerosols classes you could refer to that.

Page 11, line 7-10: Start sentence “The slight. . .” with “Summarizing, . . .” and sentence “The concomitant. . .” with “Moreover,. . .”.

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Page 11, line 14-15. This sentence is very vague and should be more specific. After inclusion of an additional table summarizing the plume results you refer to that here.

Page 11, line 16-18: Include “from these studies” after “A main conclusion”. The conclusion from Russo et al. you refer to is very general and you try to be more specific here.

Page 12: Here the authors discuss the role of “biomass burning” not clearly mentioning the important role of domestic biofuel use in Southeast Asia (see comment 1 of my Specific comments). Refer to work by Streets et al. and try to look into the $\text{CH}_3\text{Cl}/\text{CO}$ emission factor and the correlation between CH_3Cl and the anthropogenic tracer C_2Cl_4 to discuss the role of “anthropogenic” biomass burning versus “natural” biomass burning.

Page 12, line 20-23, and page 13 line 1-3: See my point 5) in the Specific comments above.

Page 13, line 4: The CO/CO_2 ratios are relatively low compared to natural fire ratios pointing to more efficient burning. This could also relate to domestic biofuel emissions which can be more efficient than “natural fires” resulting in lower CO levels.

Page 13, line 12-15. Also here the authors are too much focused on “biomass burning” from natural fires only. A higher $\Delta\text{CH}_4/\Delta\text{CO}$ could also point to emissions from the mixed usage of fossil and biofuels.

Page 13, line 16-20. See point 3 of the Specific comments. Explain in more detail the meaning of $\Delta\text{O}_3/\Delta\text{CO}$.

Page 14, line 3-17. The section on the MODIS fire map should be re-evaluated in the light of the discussion on the role of emissions from biofuel usage.

Page 15: “Therefore, C_2Cl_4 is used here as a surrogate to estimate the relative contribution of biomass burning relative to anthropogenic emissions. Slope value of 33 ppb/ppt ($\text{CO}/\text{C}_2\text{Cl}_4$) is assumed to represent the regional correlation between anthro-

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pogenic CO and C₂Cl₄. Anthropogenic CO concentration is then estimated using C₂Cl₄ concentration multiplying a factor of 33.” Include “A” before “Slope value...” and “The” before “CO concentration...” include “by” before “a factor..”. This section becomes clearer if you include a simple formula to explain how you calculate the An-CO).

Page 15: The estimate of anthropogenic CO should also be re-examined considering the fact that a part of the biomass burning CO from domestic biofuel usage is actually also “anthropogenic”. Try to explain the difference between your estimates and those from TRACE-P. Is there a seasonal difference or might there be a trend in the emissions? In any case one can assume that the fossil fuel emissions have increased significantly in Southeast Asia over almost a decade.

Page 15, Conclusions: Adjust in line with the suggested modifications.

Table 2 and 3: Use the same notification for the flights.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 21891, 2009.

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