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Atmospheric Chemistry and Physics Discussions

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.

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We would like thank the two referees for their time to review our manuscript and also for their valuable comments on our manuscript which help us greatly to improve it. Changes have been made according to their suggestions and the answers to their comments are listed as follows:

Answer to the comments from Anonymous Referee #1

General comments by referee: 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.

Answer: In the revised version, we have considered the source contribution of biomass burning, biofuel burning and fossil fuel burning to the observed pollution events. This classification is more precise and avoids data misinterpretation. The importance of biofuel is also considered and now its role during the events is discussed referring to the previous studies e.g. Streets et al. 2003.

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 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).

Answer: More comparisons have been made with results from previous campaigns, mainly PEM-West B and TRACE-P. The observed events show the occurrence and pathway of biomass/biofuel burning and their impacts on upper tropospheric air. Also, the impact of fossil fuel burning due to the rapid urbanization in this region is evident in this case study. Their relative contribution is investigated using several specific tracers. We believe this is a valuable attempt to understand the impacts of different source categories on upper tropospheric air.

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 $\Delta O3/\Delta CO$ ratios in the light of earlier studies (Jost et al., 2003; Kondo et al., 2004).

Answer: A more detailed discussion on the $\Delta O3/\Delta CO$ ratio and more comparisons

with earlier studies have now been included to clarify the origin and the age of air-masses. However, an estimation of the impact on the UT/LS is difficult to make. One would need data over longer periods.

4. Include an additional graph with vertical profiles for the 4 different ïňĆights for CO, O3, aerosols, CH3CN and acetone to elucidate the vertical structure of the plumes.

Answer: No vertical profiles have been obtained during our flights because the air intake is switched off when the flight altitude is lower than ∼500 hPa.

5. Include an additional table (Table 1) were your 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 then including W18 in table 1).

Answer: An additional table has been made to list the concentration of measured chemical species in the samples. Statistics of event and non-event samples has also been included.

Specific comments:

Page 4, line 9-10: "It is expected that in addition to anthropogenic emissions, oceanic and biomass burning emissions also inïňĆuence 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 here1) it can be expected that . . . . composition in this region."

Answer: Changes have been made accordingly.

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).

Answer: the sentence has been rephrased as "Using the chemical composition, back-trajectory analysis, cloud contact analysis and satellite images, the source region, transport pathway and the contribution of regional sources (i.e biomass burning, biofuel burning and fossil fuel burning) are investigated here."

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.

Answer: Changes has been made. The samples names are included in Fig 2 (a) and (b) with grey bars and labeled with sample names.

Page 5, line 14: Include "different" before "laboratories".

Answer: Change has been made.

Page 7, line1 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.

Answer: The mentioned mistakes have been corrected.

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

Answer: Change has been made.

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.

Answer: The earlier sentence was not clear enough. We intended to point out a non-plume-period between Event 2 and Event 3 with deceased mixing rations of chemical species. We have rephrased it as "A period without pollution with a strong decrease in

trace gas levels, except for ozone which actually increased, was then observed while the aircraft climbed to a cruising altitude of ∼11 km from 12:50 UTC to 13:15 UTC."

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

Answer: An additional table and comparison have been made.

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.

Answer: Change has been made.

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.

Answer: A more detail summary of the plumes and the measured chemical species has been presented. A comparison with pervious observation from Southeast Asia has also been made.

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

Answer: References have been added.

Page 10, line 5-7: "When air is in contact. . . .., would be expected." Include a reference

here.

Answer: A reference has been added.

Page 10, line 10-17. I find the evaluation of the δ13C results written in an disorderly and unclear manner. Please explain the meaning of δ 13C(CO2) range you find (-8.52% to -8.31%) and include a reference. Change "High CO2 values always correspond to low δ 13C(CO2). . . " to "Elevated CO2 concentrations generally correspond to low δ 13C(CO2). . . .." and try to include a reference.

Answer: we agree that this part was rather week. This part has now been rewritten.

Page 10, line 17: Include "the NMHCs" before "C H , C H ,. . . "

Answer: Change has been made.

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

Answer: This sentence was not clear and useless so it has been deleted.

Page 10, line 22-23: Clarify what "a slight increase in O3" means by including the actual increase relative to the "background".

Answer: An increase in ozone from its background (from ∼30 ppb to ∼60-90 ppb) is emphasized.

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.

Answer: "Enhancements in N12 during Event 1-4" is mentioned here to make a contrast with almost no increase in ultrafine particle N4-12 (except small increase during Event 1).

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

Answer: Changes have been made.

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 you is very general and you try to be more specific here.

Answer: The findings of previous studies now are shown in more detail with a more specific description with the focus on the different contribution in different regions.

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 CH3Cl/CO emission factor and the correlation between CH3Cl and the anthropogenic tracer C2Cl4 to discuss the role of “anthropogenic” biomass burning versus “natural” biomass burning.

Answer: The discussion on the role of biofuel burning versus biomass burning is conducted in the last part of the discussion. The importance of biofuel burning is suggested using the correlation between CH3Cl and the anthropogenic tracer C2Cl4.

Page 13, line 4: The ΔCO/ΔCO2 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 to much focussed on “biomass burning” from natural fires only. A higher ΔCH4/ΔCO could also point to emissions from the mixed usage of fossil and biofuels.

Answer: Yes. This is a possible reason for the relatively low CO/CO2 ratios. The possibility is that the plumes were impacted not only by biofuel burning but also by fossil fuel burning (Russo et al., 2003).

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.

Answer: It is indeed unfortunate that satellite images only give clues to open fires. For biofuel use, one should refer to the previous studies e.g. Streets et al. 2003 for more discussions, which has been done in the revised version.

Page 15: “Therefore, C2Cl4 is used here as a surrogate to estimate the relative contribution of biomass burning relative to anthropogenic emissions. Slope value of 33 ppb/ppt (CO/C2Cl4) is assumed to represent the regional correlation between anthropogenic 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).

Answer: Due to the vague description and obscure understanding of the estimation, we rewrote this part and focus more on the contribution of different source categories to the CO enhancements.

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 their 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.

Answer: C2Cl4 is a totally man-made chemical species and related closely to the anthropogenic activities such as industrial processes, which can only be related to the fossil fuel burning instead of any biomass/biofuel burning. It is used as a proxy to refer to the fossil fuel burning in the urban area versus the biomass/biofuel burning. The C2Cl4/CO ratio was obtained from previous campaigns during which the C2Cl4/CO

ratio in urban plumes was investigated. Therefore, we think the slope is suitable to be used for fossil fuel related CO estimation.

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

Answer: Adjustments have been made in conclusions and abstract.

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

Answer: Changes have been made.

Answer to the comments from Anonymous Referee #2

The paper reports the concurrent measurement of a range of trace gases and some limited aerosol properties from an in-service aircraft, part of the CARABIC programme. The measurements made by this experiment are highly unusual and they throw up many interesting aspects of atmospheric chemistry that are missed by very limited experiments using research aircraft. The paper is of general interest in that it adds to the inventory of chemical speciation in the upper troposphere in this region. This is in itself is very valuable since the sources of pollution in the continent are changing rapidly, and what was observed as a prevalent source or chemical distribution a decade ago many not be so today.

Overall the paper should be published but there are a number of areas which need clarification before this can happen.

1. The weakness of the analysis presented here is that it is based on a very small number of whole air samples, and their point measurement nature leads to a degree of simplification in the treatment of sources. It is a somewhat semantic point, but I struggled throughout the paper with the author's definition of each of the elevated pollution regions as being in a 'plume'. A plume has very specific meaning (to me at least) indicating a point or highly concentrated source region, being then transported, mixed and diluted. Some the elevated regions of data shown in the paper assigned as plumes cover very large distances (eg CO is elevated for 30 minutes of flight time in

plume 1); one might better refer to these as polluted airmasses 1,2,3 etc rather than plumes. If there could be some sort of clarification on this, even if it is just in definition, it would be helpful.

Answer: We have improved the manuscript in this aspect. We used "pollution event" and "airmass" to substitute "plume". Pollution event here refers to a period of time with polluted species enhancements. The air measured and studied during a certain event is called "airmass".

2. The combination of trajectory data with cloud cover is used to illustrate that convective processes could be responsible for the elevations seen from the aircraft. The approach is excellent and appropriate, but I found it very difficult to see how convincing the data was from the figures. Figures 3 and 4 show trajectories/clouds for all the flights, but what is really of interest to the reader are the trajectories and associated clouds only for those periods assigned as plumes and where there were whole air samples. I would really like to see these figures reworked so that the reader can trace backwards from the flight track and the whole air sample via trajectory to the cloud. I had to draw on the sample points from Fig 1 on to Figure 3 and 4, and then try to follow the trajectories by hand. This could be a new figure?

Answer: Figure 1, 3 and 4 have been reworked. Highlights of pollution events and whole air samples have been added. The back-trajectories during the events and the whole air sampling interval, as well as their contact with cloud now are displayed in the figures. We believe that it would be clear to show our results and better for understanding our discussion.

3. A summary table which highlighted the basic statistics for all species concentrations in and out of polluted regions would be a valuable addition to the more detailed information given in Table 1.

Answer: New table (Table 1) has been made.

4. Page 21898 lines 2-8 were highly confusing and need re-writing.

Answer: This part has been rewritten.

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