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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-range transport patterns into the tropical northwest Pacific during the CAMP^{2}Ex aircraft campaign: chemical composition, size distributions, and the impact of convection" *by* Miguel Ricardo A. Hilario et al.

Anonymous Referee #2

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The manuscript submitted by Hilario et al is a well written, technically sound analysis of the source regions and transport pathways impacting the airmasses sampled during the recent NASA CAMP2EX campaign. Statistical analysis of back trajectories from a meteorological reanalysis, together with precipitation satellite products are used to assign source regions and evaluate the possible impact of wet deposition/convection on these pathways. Several sensitivity analysis are performed to probe the robustness of the assignment, in particular in the vertical. Overall, with this approach the authors are able to assign a unique source region to 1/3 of airmasses sampled. The (fairly

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limited) gas and aerosol instrumental package on the NASA P-3 is then used to support the regional assignments made, and to illustrate the effects of vertical motion and wet deposition upon these variables.

While the authors imply in the abstract and introduction that this work is highly relevant to the general interpretation of atmospheric chemistry measurements in the Equatorial Pacific, the synoptic results presented are really only relevant to the time of year of CAMP2EX (Sep/Oct), they are based on a heavily biased sample (by the flight plans of the research aircraft involved), and with only 33% of positive identification in that biased sample it seems hard to justify calling them representative for the region at large. The discussion of vertical transport and source mix is interesting and well written, but given the limitations of the payload they can at best confirm expected trends based on our current understanding. These results might set the stage to much more novel results gathered during CAMP2EX, but those results are clearly not part of the paper as written.

So to summarize, this is a very good description of the synoptic conditions for a particular subset of CAMP2EX measurements. Hence, I do agree with the other reviewer that, as written this would make a fine ACP measurement report and I would strongly support resubmission as such, once my detailed comments are addressed. Alternatively, the authors could shorten the manuscript significantly (lots of technical details could easily go into the SI to keep it at a reasonable length) and add some of the science that they propose in their next steps, and resubmit this as a research article.

Major comments: - This manuscript uses as lot of acronyms, many of which are fairly specific to this study/meterological context, which makes it at times fairly hard to follow. So I would suggest to include a simple lookup table, especially for the benefit of casual readers. - The whole discussion on the species ratios is very qualitative. While the authors emphasize a few times that they have very little information on the sources and sinks along the trajectories, the same is true for the emissions themselves, as currently written. The discussion would be considerably stronger if the authors had compiled

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average emission ratios for each source region based on regional/global emission inventories (e.g. REAS2, CEDS), and comparing those numbers to the ratios actually observed. That would not only put the analysis on a more quantitative basis, but also make clearer how much contrast to actually expect for different air masses. In that context, I would encourage the authors to use one consistent denominator for emission ratios. E.g. while indeed peat burning has higher sulfate emissions than other fuels (Line 410), relative to fuel burnt (or CO), looking at the SO4/OA ratio is not really helpful, since we expect dOA/dCO to be higher in this case and dSO4/dCO lower. Using a consistent metric (also in Fig 7) would streamline the discussion and make quick comparisons easier. - On a similar note, the discussion of the size distributions (Fig 8) is fairly speculative and confusing (at least to me). The difference between clean FT and BL is clear cut and obvious, and does confirm the analysis of the vertical distribution by species/species ratio. But for the BL data, the current discussion focuses too much on possible differences in the source region and too little on processing, which especially when discussing the number distribution is a major concern. E.g. size distributions in large urban areas (so primarily EA) do typically have a large Aitken mode composed of fresh HOA/nitrate (e.g. see Zhang et al, 2005), but that smaller mode is short lived due to coagulation/aging and typically is not observed downwind of the urban core, let alone 96-120 h downwind, so I would be careful interpreting the MC/EA differences in this way (e.g most of this could be secondary sulfate from SO2 oxidation downwind, but again, without supporting evidence, this is just a guess). Similarly, the dip between Aitken and accumulation mode in the MBL has long been attributed to cloud processing (Hoppel et al, 1985), so trying to interpret the depth of that dip based on the source region seems like a stretch. Lastly, given the sampling region, the reported AMS + BC speciation may or more likely may not fully represent the total aerosol volume, hence complicating the interpretation further. So I would significantly shorten that part of the discussion, which without considerable additional information is just not well supported. One possible way to improve on the current discussion would be to map the AMS mass size distributions (should there be any and their S/N of high

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enough quality) on the volume size distributions (Fig S9). E.g. if there are significant differences in composition at 80-120 nm between different regions, that would certainly be an interesting result and could give more insight into the processes involved. But given the low overall concentrations, this might be challenging. - Section 2.3/Figure 9: Using satellite data instead of e.g. the NCEP reconstructed meteorology along the back trajectories is a sensible choice, since the higher spatial resolution of most of these satellites products can be beneficial. However it is unclear as written how this four data products are merged and prioritized, especially given the very different time resolution of the individual products. This does not matter for the bulk of the analysis (where as Table 1 shows, the results for all approaches are compiled and give consistent results, with scatter roughly proportional to the fineness of the temporal resolution, as expected). But it is unclear which approach or combination of approaches went into the X-Axis of Figure 9a for the individual points shown, please elaborate. Related, it would be actually very useful to compare the different APT factors with the calculated convective and precipitation probability in the meteorological reconstruction used for the back trajectories. The performance of these reconstructions has steadily improved in recent years, so a brief assessment on how different the results would be if only relying on the model would be a valuable addition. - The scavenging mechanism for MC air in the FT is an interesting theory (also pretty speculative, as pointed out by the authors). However, the discussion does not sufficiently acknowledge that it's particles that are scavenged, not AMS species. And as the size distribution data shows, there is not much evidence for external mixing of sulfate and OA for MC air in particular (again, AMS SD data would be helpful here). So scavenging is unlikely to be species specific. In addition, BBOA tends to age fairly fast, especially in a high OH environment such as the tropics. I would assume that AMS f60 is fairly low/almost gone by the time these airmasses were sampled. If so, that would suggest a fairly high AMS f44/ O/C ratio. Which in turn means that a) those particles are likely internally well mixed (see e.g. Gorkowski et al, 2020) and b) the OA is reasonable hygroscopic (e.g Brock et al, 2016). So again, this makes the fact that OA is not scavenged as well as sulfate fairly

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puzzling and needs to be discussed in more detail. - I am not convinced that Fig 9b shows what the authors intend. My reading of Fig 9a is that to a large extent, the trend in dBC/dCO is driven by different emission ratios for each region (not all of it, which is why this is a useful figure). But showing the data in the aggregate as done in Fig 9b averages again over these differences and will be hence just reflect changes in source region, not wet deposition

Minor comments: - Abstract: As noted above, this study does advance our understanding of long range transport in the TWNP, but it does not show that most of this applicable outside the intensive CAMP2EX period. Hence I would disagree that this research can "...[guide] international policymaking on public health and climate" (also end of the Introduction) and would suggest rephrasing. - Intro, 2nd paragraph: Most of the chosen names for the source regions are not standard by any means, they are just good operational monikers chosen by the authors for this particular study. And in fact, they are introduced as such later (with coordinates), L105-L107. So I would suggest to the authors to just use country names at this stage, and refer to the operational definitions later (so e.g. "Korea and SE China, referred to as EA in the following). - Line 101: Given the environment and aerosol mix, I think the recent paper on convective scavenging by Yu et al (2019) is relevant as well: Yu, P., Froyd, K. D., Portmann, R. W., Toon, O. B., Freitas, S. R., Bardeen, C. G., Brock, C., Fan, T., Gao, R.-S., Katich, J. M., Kupc, A., Liu, S., Maloney, C., Murphy, D. M., Rosenlof, K. H., Schill, G., Schwarz, J. P. and Williamson, C.: Efficient In-Cloud Removal of Aerosols by Deep Convection, Geophys. Res. Lett., 46(2), 1061–1069, 2019. - Line 123: Please indicate the refractive index the optical calibration of the LAS is based on. For the uncertainty, please provide a reference or describe the uncertainty sources in more detail. Also, I would note that while the FIMS and SP2 have similar upper size limits, 1000 nm optical size (again, depending on the calibration used) is under most conditions more than the D50 for the AMS equipped with a PM1 lens, so at a minimum I would suggest adding the size ranges for each instrument and clarifying how 1000 nm Dopt compares to those. - Line 129: deCarlo et al (2008) is a field study (and neither the first AMS flight de-

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ployment). The best instrumental reference here is likely Canagaratna et al (2007) (for the AMS in general) or deCarlo et al (2006) (for the HR-AMS in particular, if this is indeed the instrument flown, please specify). Please include this information. - Line 133: It is unclear how the detection limits were estimated, are these from blanks or using the method from Drewnick et al (2009). Importantly, detection limits are only meaningful in combination with their sampling period. I assume this is for 30 s, based on the discussion, but please indicate. - Line 134: It is unclear what this statement is supposed to mean. Data under DL at e.g. 30 s sampling time does still contain information that can become meaningful when averaging for longer periods. If the data was rather zero'd, this would actually bias the data. Can the authors clarify? - Line 135: Again, please provide a cite/justify the uncertainty estimate. - Line 137: I would suggest adding an entry to Table 1 with the AMS totals excluding RF9 - Line 142: While this is a sensible choice for the optical instruments, the tropical pacific is very cloudy. Can the authors provide numbers on how this affected the data coverage? And how exactly was "cloudy" defined? - Line 159: While discussed later, convection along the trajectories should be mentioned here as well - Line 188: Isn't case 1 equivalent to e.g. long range transport from farther away source regions (e.g India, West Asia). If so, I would suggest making that explicit (also in the context of evaluating the impact of LTR on the CAMP2EX domain). It would also be informative if you could provide (even rough) percentages for cases 1-4. - Line 321: It is unclear what "unimodal" is supposed to mean here - Line 345: In the absence of better tracers, this is probably the best approach. How much data was screened this way? - Line 353: The altitude profiles are fairly coarse. A higher resolution altitude profile of temperature and RH would be better to establish the most sensible BL height for the sampled data. - Line 398: Again, in the absence of source emission ratios this is not very convincing. The marine continent has many biogenic and anthropogenic sources of CO2 and CO that will contribute to the total. It is possible that the fire emissions overwhelm all that signal, but the authors have not made that case yet, so it is at least as likely an explanation as the ones they mention. - Figure S9: It would be much better to plot this with varying

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y-scale and the total volume mentioned in the legend, it is very hard to actually see the differences for the cleaner sectors - Author contributions: Please be more specific on the individual contribution of each author, per ACP policy.

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