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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An overview of the Amazonian Aerosol Characterization Experiment 2008 (AMAZE-08)" *by* S. T. Martin et al.

S. T. Martin et al.

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We appreciate the reviewers' careful reading of the manuscript and their valuable comments that have led to improvements in the manuscript. The reviewer comments, our replies, and our updates to the manuscript are sequentially listed, as follows: 1. The Hatch-Choate relationships are not defined or referenced, and it is not clear what is meant by saying that the "parameters for the surface and volume distributions satisfy" the relationships. They must, since they are lognormal descriptions of the data, and the Hatch-Choate relationships are defined for lognormal distributions.

* This is a good point. In explanation, we calculated the surface and volume distribution from the measured number distribution. The bimodal lognormal fit was done to the surface distribution and the resulting fit curve was converted into number and volume

by the Hatch-Choate equations. By this method, we obtained a consistent data set that matches all three moments of the size distribution.

* The text is revised to read: "As a consistency check, the parameters for the surface and volume distributions satisfy the Hatch-Choate relationships to the number distribution."

* A citation is added for the Hatch-Choate relations: "Chapter 4 of W.H. Hinds, Aerosol Technology, Wiley: New York, 1999".

2. In the following 2 sentences, the goodness of closure is defined as the "residuals of log10 values". I don't know what this means, and an equation (or at least a reference) would be extremely helpful.

* The original text was confusing. The revised text states: "The bottom panel of Fig.~12 shows the log$_{10}$ residuals between the measurements and the parameterization."

3. In the next sentence (lines 5-7), "the parameterization is constrained within 50% of the measured median". Does this mean than the parameterization and the median agree within 50%, or was there some constraint in the fitting process?

* We agree that the use of the word "constrained" was not informative, and we removed it from the revised text. The revised and clarified sentence reads, "Across the full size range of 20\,nm to 3\,\unit{\mu}m, the median residual of the parameterization is within 50{\%} of the measured median."

4. Figure 12, which accompanies this section, shows number, surface, volume, and residual plots, nicely displayed. However, I am not at all in favor of the logarithmic ordinate. It masks the proportional contribution of each size class to the total number (or surface, or volume). The objective should not be to "show all the data", which a log-log plot allows, but to show which data are important to that moment. Furthermore, the area under the curve is not meaningful in a log-log plot, while it is in a lin-log plot.

* The reviewer has a valid point of view, and we do not disagree with it. Nevertheless,

a choice must be made about how to present a figure and toward what purpose. The reviewer's purpose, which is a visual representation proportional to the size of each class, can be prepared by future interested readers by use of the size distribution parameters provided in Table 3. Our purpose, however, was different than this goal. We want to give the reader a full perspective of the size distribution from 20 nm to 3 um in a single plot, and the log y-axis scaling is necessary to this purpose. A linear y-axis would only show the particles having diameters < 300 nm, which make up 99% of the particles by number. The presence of larger particles, however, even at small number fraction, is of importance in the context of primary biological particles and their role in the atmosphere. For this purpose, the log-log plot is informative.

5. Also in Fig. 12, there is a big discrepancy for particles with diameters > 2 um between the APS and the OPC. The OPC shows a remarkable coarse mode extending beyond 10 um, while the APS shows a peak near 3 um and declining concentrations for larger sizes. These are very substantial differences, and one would arrive at two different conclusions regarding the magnitude of the coarse mode from these 2 measurements. Which is correct? How big are the uncertainties for diameters > 3 um? Clearly the OPC data do not fit the bimodal lognormal model presented as a parameterization to be used for modeling, so this issue needs to be discussed.

* The reviewer's comment on this topic is well taken. The manuscript states:

* "The distributions for these larger particles are based on the UV-APS and OPC measurements. The figure shows that the OPC observes the presence of particles with sizes larger than 4 μm that the UV-APS does not indicate. The explanation may be that the aerodynamic cutoff of the inlet-sampling system was between 5 and 7 μm, consistent with the size distributions observed by the UV-APS. The implication is that the particles detected up to 10 μm with respect to optical diameter (which represents the efficacy of light scattering compared to the calibration particles of polystyrene latex spheres) have smaller aerodynamic diameters. For instance, these results can be explained by non-spherical particles having a dynamic shape factor above unity, parti-

cles having a density less than that of water, or a refractive index greater than that of PSL material. For the typical particle types observed in the Amazon Basin, a dynamic shape factor larger than unity is perhaps the most plausible explanation (e.g., primary biological particles such as fungal spores)."

6. In section 3.2.2, page 18158, beginning line 9, there is a discussion of how the fractional abundances of PBAP markers compares to laboratory results, producing an upper limit of 5% primary particle contribution toward the accumulation mode. This is an important point, yet the data supporting this statement are not presented. Could this be shown graphically, or at least expanded upon a bit with references and some more meat so that we understand how this finding, which contradicts past published reports, is determined?

* The reviewer's query is well placed. A manuscript by Schneider et al. is under preparation, and this manuscript will present the details of the analysis and the results. At present, results are also available in the thesis: Freutel, Friederike: Identifizierung charakteristischer massenspektrometrischer Marker für primaere biologische Aerosolpartikel, Masters Thesis, University Mainz, 2009 (in German).

* Citations are added to the manuscript both to the thesis and to the manuscript under preparation.

* Inclusion of a significant section on this important topic in the AMAZE-08 overview manuscript is beyond its scope, both because this section must be long to be meaningful and because it is (therefore) the topic of a forthcoming detailed manuscript.

7. Page 18158, lines 25-28. The reconciliation of past PBAP measurements and the current findings is the size being evaluated. For the present study, the diameter is given as 0.06-0.8 um. Is this vacuum aerodynamic diameter, in which case the true diameter range might be $\sim$0.04-0.55 um? A quantitative discussion of the AMS size classes is needed in the measurements section, since this ends up being an important part of one of the key findings of the paper.

* Yes, as the reviewer states, for the AMS the size range is expressed in vacuum aerodynamic diameter. For use in conversion to a volume-equivalent diameter, the measured density is 1400 kg mˆ-3 for AMAZE-08, as reported in Gunthe et al. (2009): Page 7556: S.S. Gunthe, S.M. King, D. Rose, Q. Chen, P. Roldin, D.K. Farmer, J.L. Jimenez, P. Artaxo, M.O. Andreae, S.T. Martin, and U. Poeschl, "Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity," Atmospheric Chemistry and Physics, 2009, 9, 7551-7575

* For PBAP, the conversion to a volume-equivalent diameter is less well constrained because these types of particles are typically not spherical. They therefore have no “true” diameter and can only be described in terms of equivalent diameters, such as the aerodynamic diameter in Graham et al. (cited page 18158, lines 25).

* We agree with the reviewer’s intent and desire to have more refined size distributions, and the call for doings so is a welcome suggestion. Nevertheless, given the considerations of non-spherical PBAP particles as well as possible scaling factors for the AMS, more precise size information than provided in the manuscript would be promising more information than we can deliver. That said, the uncertainties in size are on a scale of order unity, whereas the cited lines 25-28 refer to a much larger separation that is several times unity, i.e., 2.5 um compared to 60 to 800 nm. We judge that the written conclusion (lines 25-28) that PBAP particles contribute toward the 2.5 um end of the size range rather than in the accumulation mode is an informative and reasonable one, without getting too detailed about the exact "true" size of the demarcation. Figure 1 of Poeschl et al. 2010 reinforces this conclusion. In the revised manuscript, an additional citation is added to Poeschl et al. to direct the interested reader to more detailed information.

8. Section 3.2.3, p. 18159, line 25. There is an opportunity here to compare results with the non-biomass burning, non-dust cases from the AMMA campaign, for example Capes et al., Atmos. Chem. Phys., 9, 3841-3850, 2009. I am struck by the consis-

tent, relatively low amounts of SOA produced over both the African and Amazonian rainforest. This suggests a very short lifetime in the rainy season.

* We agree with the reviewer that comparisons between the AMAZE-08 and AMMA campaigns is helpful. The revised text include the following new material: “These results for the Amazon tropical rain forest can be compared to other recent observations over tropical forested West Africa during the wet season (Capes et al., 2009) as well as measurements above a maritime tropical forest in Borneo, Malaysia (Robinson et al., 2010). In all measurements, the organic mass concentrations (ca. 1 ($\pm $30{\%})) are consistently lower for the tropical rainforest compared to temperate coniferous forests (Zhang et al., 2007). The three tropical locations also have in common that secondary organic material is identified as the major organic component in the submicron mode.”

* Capes, G., J. G. Murphy, C. E. Reeves, J. B. McQuaid, J. F. Hamilton, J. R. Hopkins, J. Crosier, P. I. Williams, and H. Coe (2009), Secondary organic aerosol from biogenic VOCs over West Africa during AMMA, Atmos. Chem. Phys., 9, 3841-3850. * Robinson, N. H., et al. (2010), Evidence for a significant proportion of secondary organic aerosol from isoprene above a maritime tropical forest, Atmos. Chem. Phys., Submitted. * Zhang, Q., et al. (2007), Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, Geophys. Res. Lett., 34(13), L13801, doi:13810.11029/12007gl029979.

9. Section 3.2.4, p. 18161, lines 17-19. The suggestion is made that the volatility spectra from AMAZE are consistent with SOA formation, while those from polluted environments (California and Mexico City) are consistent with POA emissions. I think this is a misstatement of the findings from polluted regions; most of the organic mass there is SOA. It may have a different structure, and thus a different volatility distribution, than the biogenic SOA in AMAZE-08, but it is largely secondary nonetheless.

* We agree with the reviewer that the words used in text were confusing. The inten-

tion was to emphasize that there are important differences between AMAZE-08 and SOAR/MILAGRO with respect to particle sources and that these differences can influence the thermodenuder observations. For instance, in MILAGRO, there was a substantial contribution of primary emissions, along with biogenic and anthropogenic secondary sources, to the atmospheric aerosol. In AMAZE-08, the sources of the organic material were much more uniform, i.e., dominated by secondary organic processes. Each of these types of organic material (i.e., biogenic secondary, anthropogenic secondary, and primary) has different volatility, with biogenic secondary material observed as most volatile in many cases.

* The clarified text is as follows: "Urban primary and secondary organic material (i.e., having a high fraction of anthropogenic precursors) is less volatile than biogenic secondary organic material (Huffman et al., 2009a,b). The lower volatility of SOAR/MILAGRO compared to AMAZE-08 might also be explained in part by the presence of inorganic and black-carbon components that are commonly internally mixed with organic material in many urban areas, in contrast to the organic-dominated composition of individual particles in the Amazon Basin."

10. p. 18142, line 3. "conveyor belt" is confusing terminology, given the meteorological phenomena of "warm conveyor belts" etc.

* "Conveyer belt" is eliminated and replaced by "advection".

11. p. 18146, line 23. Was this a tethered balloon, and if so, what was its altitude range?

* The information is provided in the revised text (i.e., tethered and up to 800 m).

12. p. 18147, line 11. A switch was made from Celsius (earlier in the text) to Kelvin.

* The correction is made.

13. p. 18147, line 21. Was the inlet cutoff calculated or measured?

* The value was calculated based on known dimensions and flow rates through the sampling lines. The text is revised to make this point clear.

* The upper-end cutoff was also empirically confirmed on one occasion by making OPC measurements at the top of the tower simultaneously with measurements at the termination of the sampling line in the container.

14. p.18148, line 20-21. Was there any condensation possible in the gas sampling lines in the 23C trailer (could be slightly cooler than ambient)?

* The top of the tower was consistently cooler than the temperatures inside the instrument container. Based on the time series of relative humidity and temperature inside the container and at the top of the tower, condensation in the gas sampling lines was not expected inside the container for >95% of the measurement time.

15. p. 18152, line 1. Figure S4 was not readable in the PDF; all other .figures were fine.

* In clarification, Figure S4 is a time series plot of primary data. The intention is that this figure should be viewed on-screen at 400% zoom with use of the arrow keys for left and right scrolling within Adobe Acrobat. The aspect ratio of the figure is not suitable for printing to paper. The caption to Figure S4 is revised to include these instructions.

16. p. 18179. Add list of acronyms for "organization" at bottom of table.

* The organizations are presently listed in the caption of the table instead of at the bottom. We support the reviewer's recommendation, and we will explore with the typesetting editor the flexibility of "ACP Style" with respect to this suggestion.

17. p. 18185, Fig. 4. Would you consider adding a regional map showing the same trajectories at a scale to see the regional transport patterns vs. location of cities?

* The reviewer highlights Figure 4, which is on the scale of the Atlantic Ocean and has the purpose of showing ten-day backtrajectories. The reviewer has a good suggestion

to also scale-in to explore regional effects. Figure 8 shows these patterns within the Amazon Basin as an overlay on fire maps; the purpose of the figure is to consider possible influences of regional biomass burning. An even tighter map for the transportation routes of the city of Manaus and local meteorology is not provided; instead, the data have been screened to remove pollution events that are at times clearly evident from the transitory spikes of black carbon, particle number concentration, and NOx. The text discusses the screening for these pollution events.

18. p. 18186. The figure caption does not describe the wind roses adequately. Do the rings indicate fraction of time (e.g., 0.1, 0.2, 0.3) in each sector?

* Yes, the reviewer's description is correct. The caption is accordingly updated.

19. p. 18192, Fig. 11. The black line (PBL top) is described in the text but not in the figure caption.

* The figure caption is revised according to the reviewer's suggestion.

20. Page 10 session 3. Observations and findings – The months of February and March correspond to the wet season in the Amazon. Day to day satellite images show that cloudiness is a constant feature sometimes organized in large westerly travelling systems and sometimes more local systems (Greco et al, 1990, Cohen et al 1995). In Machado et al 1998, specifically in their Fig. 5c, it is seen that during the wet season (DJF) cloud systems have lifetimes of about 12 hours and travel due west. The point here is that the synoptic trajectories represent averages over a large amount of vertical recycling that goes on in convective clouds as soon as the air mass enters the northern coast of South America and over the 1600 km of pristine forest until reaching the AMAZE08 experimental sites. In the Tropical Atlantic the air mass originated in Africa has to go through some recycling also due to convection in the ITCZ. In the approximate 10-15 days that it would take to go from Cape Verde to the Amaze08 site, the air mass might have gone through vertical loops in perhaps half the number of days. As reported by Andrea et al 2001, the convective trajectories of air can have some

unexpected results as updrafts and detrainment heights provide a pathway for the low level air mass to reach different heights, with differing synoptic wind directions. Freitas et al 2006 show that the plume rise from vegetation fires in the Amazon can be ejected at different heights thus providing an explanation for the layered structure often seen by LIDAR in the region. Page 18149 lines 13-14, "predominantly and consistently", this is an average that is continuously disrupted by convective cloud systems and associated up and downdrafts.

* The reviewer brings up important points regarding one of the "big" question in atmospheric sciences, specifically how valid is a Lagrangian parcel model as an approximation of atmospheric transport. The specific statement in the manuscript mentioned by the reviewer is: "The figure shows that the synoptic-scale winds came predominantly and consistently from the northeast, travelling across the Atlantic Ocean and then over 1600 km of nearly pristine forest before arrival at the research site. The low-altitude flow was from the northeast, changing to easterlies at mid-altitude." This statement as written is in itself accurate. However, in line with the reviewer's analysis, this statement does open itself to the charge of "error by omission", i.e., a reader is presumed to have prior knowledge about the importance of vertical transport, and a reader who does not have this prior knowledge might read the paper and form an incorrect impression that transport is entirely advective. In this regard, we have revised the text to make these points clearer (see further below for updated text).

* As a scientific matter, AMAZE-08 was carried out in the trade wind belt, where a Lagrangian parcel mode is a fairly legitimate approach. The typical trade wind convection, which happens between North Africa and the east coast of S. America, is relatively shallow, and it results in the mixing between a marine boundary layer and smoke- and dust enriched layers reaching up to 5 km. Given that there is no strong shear between the surface and 3-5 km, convection in this system results in dispersion, but not a serious violation of a mean-flow assumption. The validity of using the mean-flow approach is shown very convincingly in the Ben-Ami et al. (2010) paper cited in the manuscript

and that was part of AMAZE-08.

* Deep convection is not very much an issue with an interpretation of the results of AMAZE-08. It results in venting of some of the tracers from long-range transport to the free troposphere and a replacement of the vented volume by free troposphere air, leading to a dilution of the tracers. These aspects do strongly perturb the upper troposphere, as shown in Andreae et al. cited by the reviewer. For the AMAZE-08 measurements, however, which sampled the lower volume of the atmosphere, the dilution does not contradict our statement that airmasses in the Amazon are influenced by long-range transport from Africa. The preservation of aerosol layering, representing contiguous transport from Africa to the S. America and therefore incomplete vertical mixing, is also strongly supported by the LIDAR publication by Ansmann et al. 2009 (cited in manuscript) as well as an addition manuscript of Baars et al. (in preparation; presented at several scientific meetings already).

* During AMAZE-08, the ITCZ was south of the study area.

* Taking all of the above points into account, the following is added to the updated text: " The trajectories represented in Fig.$\sim$4, suggesting smooth trajectories of air parcels, are a HYSPLIT product that uses a Lagrangian framework and omits treatment of vertical transport. Treating atmospheric transport over a ten-day period with a Lagrangian parcel model is a strong simplification of the complexities of atmospheric movement. Vertical exchange and cloud processing take place along the path of transport and modify concentrations and properties of airmass tracers. Nevertheless, the consistency of the trajectories from day to day and the weak vertical shear in the lower 3-5 km of trade winds both suggest that the representation in Fig.$\sim$4 is a reasonable first-order description of airmass transport. In the particular situation of AMAZE-08, there is strong observational support for the validity of this approach presented in the study. Ben-Ami et al. (2010) documented the transport of dust and smoke across the Atlantic Ocean along the path suggested by the trajectories in Fig. 4 by using a combination of remote sensing and surface observations."

21. Complete paragraph (page 18149, line 20ff). The long range transport is a possibility but with some uncertainty. Local sources to the northwest of the region may have to be taken into account as they not necessarily associate to fresh biomass burning material if the material has had a lifetime of several days until reaching the experimental site (see Freitas et al 2006).

* The reviewer brings up a good point about the possibility of local sources of biomass burning. During the data analysis and science meetings, the AMAZE-08 investigators had a combined deep look at this topic from our available data. The purpose of Fig. 8 in the manuscript is to emphasize that trajectories reaching the research site did not pass through the biomass burning to the northwest of Amazon Basin. Within the data sets themselves, both the PTR-MS measurements of the gas phase and the AMS measurements of the particle phase did not detect tracers at levels or in ratios that suggested biomass burning had an influence on the measured air masses within the time scale of several days (i.e., the approximate residence time of air in the Basin).

* More specifically, from the PTR-MS data, the maximum benzene/acetonitrile enhancement during AMAZE-08 was 0.25 ppbv/ppbv. For comparison, from the TROFFEE campaign that investigated biomass burning in the Basin, a ratio of 1.2+/-0.8 ppbv/ppbv was measured (Karl et al. 2007). The calculation based on 10ˆ6 OH cmˆ-3 is that the minimum time (i.e., benzene_0/ACN_0 > 0.4) since exposure to biomass burning is 4.5 days. The expected time (i.e., considering the midpoint of measurements instead of uncertainty extreme) is 15 days (i.e., benzene_0/ACN_0 > 1.2).

* The absence of elevated signal intensity for biomass burning markers in the AMS (i.e., m/z 60 and 73) above their background levels (e.g., Docherty et al., 2008) suggests that fresh biomass burning made a negligible contribution to organic particle mass concentrations during AMAZE-08. The AMS markers are reduced with age on a timescale of several days, so biomass burning after transport from Africa would not contribute to the marker intensity.

* The updated text reads: “In contrast, an influence of regional biomass burning appears less plausible because of the absence of fires along the in-Basin trajectories (Fig.∼8). The absence of markers of fresh biomass burning both in the particle-phase mass spectra (e.g., elevated signal intensity at \textit{m/z} 60 and 73) and in the gas-phase mass spectra (e.g., the benzene:acetonitrile ratio) further confirms this conclusion (Chen et al., 2009; Karl et al., 2009).”

* Karl, T., Christian, T.J., Yokelson, R.J., Artaxo, P., Hao, W.M. and A. Guenther, 2007: The Tropical Forest and Fire Emissions Experiment: Method evaluation of volatile organic compound emissions measured by PTR-MS, FTIR, and GC from tropical biomass burning, Atmos. Chem. Phys., 7, 5883-5897

22. Page 22 – reference to recently published Poschl et al 2010 Science paper

* The citation is added.

23. Page 27 – lines 596-605 – I am not sure I follow your reasoning. What exactly is considered a good agreement with ground observations? The statement “suggest that precipitation from cold-cloud processes can be expected with progressive frequency for cloud temperatures of -20C and colder” is something that should be expected even without these new measurements. So what is really new?

* A direct relationship between cloud top temperature and precipitation is not yet fully established. Hanna et al. [2008] examined the correlation between satellite inferred cloud-top temperature and precipitation over the contiguous United States and found varied relationships depending on precipitation type (snow, rain, freezing rain, and sleet). For example, although freezing rain often occurred for cloud top temperatures of -8 °C, they found virtually no cases during winter during which snow formed at temperatures warmer than -15 °C. These authors did not have access to any ice nuclei measurements and had to conjecture about the absence of a sufficient number of IN to explain the observations for snow. Our findings, which include surface IN measurements, are similar to those of Hanna et al., but the new findings apply to a wet-season

remote tropical climate. Our ground-based measurements indicate that fewer than 10-3 IN cm-3 are active at T > -20 °C, and, combined with the estimate of Fletcher [1961] of the number of IN needed to initiate precipitation, our data suggest that cloud top temperatures must reach temperatures of -20 °C or colder for precipitation initiation in this region. This is consistent with the concurrent MODIS observations, and to our knowledge this is the first study that has combined ground based IN studies and satellite data to constrain the link between IN number and cloud glaciation.

* The text has been modified to include this additional information and perspective: “These results are similar to those reported by Hanna et al. (2008), who examined the correlation between satellite-inferred cloud-top temperatures and snowfall patterns over the contiguous United States. Snow did not form for temperatures warmer than $-$15\,{\degree}C. To explain this observation, Hanna et al. suggested that there was an absence of sufficient IN.”

* Fletcher, N. H. (1961), Freezing nuclei, meteors, and rainfall - Do tiny particles from meteor streams influence rainfall over the earth’s continents?, Science, 134, 361-367.

* Hanna, J. W., et al. (2008), Cloud-top temperatures for precipitating winter clouds, J. Appl. Meteorol. Climatol., 47(1), 351-359.

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