

***Interactive comment on “Aerosol particle
properties in the tropical free troposphere
observed at Pico Espejo (4765 m a.s.l.), Venezuela”
by T. Schmeißner et al.***

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Author response to Anonymous Referee No. 1

Thank you for your comments to the manuscript. For clarity, your statements will appear in italic face, and our response in standard face.

We were able to account for all your comments, see below.

“Structure of the manuscript: I find the manuscript rather hard to read. Most of the Result section (Section 3) remains entirely descriptive. Features of the observed data series are described in very detail, but their interpretation is then only given in Section

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4. *The reader-friendliness of the entire manuscript will in my opinion be strongly improved by discussing the data at the same time as the data are presented. Therefore I strongly suggest combining Sections 3 and 4.*"

Since it is really a bit hard to read measurement results in one section and the discussion of these results in a different section we merged both sections together into one named "Results and discussion". Now observed characteristics or features of aerosol number size distributions for the different FT regimes and seasons are discussed immediately.

"Data included in the study: It is mentioned that also an aethalometer was operated at the site (and maybe also further aerosol instruments?). I understand that this manuscript focusses on particle number measurements. Since an influence of biomass burning on the measured size distributions is discussed in the manuscript, any additional information gained from the aethalometer should however be presented here as well."

There is no aethalometer installed at the measurement site, but a particle soot absorption photometer (PSAP). Data from the PSAP is not fully analyzed yet since data progressing is always connected with an enormous expenditure of time. Furthermore, it is planned to use this data in a separate paper. Moreover, the main focus of our work is not on biomass burning and the nature of the aerosol sources, but on the analysis of the aerosol number size distributions with regard to a seasonal variability.

There is no further aerosol instrumentation on the site, but other devices: besides the automatic weather station, there are two webcams installed producing images from the station facing the southern and northern slope every 15 minutes. In-situ ozone concentrations are also measured using an Ansyco (UV) Photometry Ozone Analyzer (model O341M, S.A.) with UV-irradiation at 254 nm and a detection limit of 0.4 ppb. An analysis of these ozone measurements was already published by Calderón et al. (2008).

"Sampling conditions: The authors state that under cloudy conditions, the examined

aerosol consists of the interstitial aerosol plus an undefined number of cloud drop residuals. Although data with $RH > 95\%$ were excluded (used as cloud criterion, see also next comment), at least an estimate of the inlet cut-off would improve the data quality assurance. Furthermore, to what extent do these residual cloud droplets dry off in the inlet?"

The cut-off of the whole sampling system is estimated to be about $10 \mu\text{m}$. The limiting factor with regard to the sampling of large particles is in our case not the inlet, but the division of the sampling line among the instruments inside the station. During cloudy conditions we do not exactly know what the real cut-off is as it is defined by the efficiency of cloud droplet sampling.

Droplets which enter the sampling line dry off completely, since the RH of the sample air inside the station prior entering the instrumentation is between 10 and 20%. All this is now mentioned in the revised text.

"Cloud definition: In the presence of a patchy cloud, I would expect a variability between 90 and 100 % RH during an individual DMPS scan (15min), which would a) influence the performed categorization of this size distribution (cloud/no cloud) and b) the size distribution itself. Do the authors have any idea on the density of the clouds which were usually be present at the site? In case of frequent patchy clouds, the above effect may influence the results. This needs to be addressed."

Yes, there are in deed patchy clouds at the sampling site. We are aware of the problem that these clouds might not be fully considered by our method. We are also aware of the fact, that the occurrence of patchy clouds and subsequent wrong classification of data measured under such conditions might have an influence on the size distribution of this data. But since the paper focusses mainly on long-term trends and a large number of data is averaged, the impact of these effects on our results will be rather small.

Moreover, we tested the filtering algorithm also for lower RH-values (down to 90%) and

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could not observe a significant difference. But you are right, this issue needs to be addressed in the paper and we have done this in the revised version.

“Section 3.1: The season-dependent average number concentration (plus standard deviations) are given in the text and in Table 3. This is redundant, I suggest skipping the Table. Also, Figures 2 and 3 basically show the same information. I suggest showing only Figure 3 (the box plots). The average concentrations plus standard deviations are also prominently listed in the Abstract. Based on the large standard deviations the question may arise while reading the Abstract whether the reported differences are significantly different at all on a statistical basis. Although the box plot in Figure 3 nicely shows the differences, this issue should be taken into account to avoid confusion.”

It is true, Figure 2 and 3 basically illustrate the same issue. Therefore we followed your advice and skipped Figure 2 and only leave the Box-and-Whisker plots in the paper. Regarding Table 3, we changed the standard deviation against the standard deviation of the mean value. This produces not such large and confusing numbers. Moreover, we consider these values as quite important since they present the seasonal variability of the Pico Espejo aerosol in a very illustrating way. Therefore we decided to keep the Table in the paper.

“Section 3.2 / Data base: Size distribution analysis is only shown for 2008. What about the data from the rest of the measuring period?”

Yes, we first focused only on the year 2008, but of course you are right, we should use the whole amount of data. For the revised paper we therefore made this size distribution analysis for the several months of the complete measurement period (i.e. for each month from March 2007 until March 2009) separately and applied the mode fit procedure on it. Afterwards we created mean size distributions for each month of the year (for example for March consisting of data from 2007, 2008 and 2009) and applied the mode fit procedure on these mean size distributions. Therefore, we could see that extraordinary features were reproduced every year and are thus in deed characteristic

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features of the Pico Espejo aerosol. For the revised paper we then decided to show the data averaged over the course of one year and not each month of the complete measurement period separately.

“Section 3.2 / Distribution fitting: I see a general problem with the automatic fitting of three modes into every individual size distribution. While the fitting program will always return a mode diameter value for all three modes, these numbers do not physically make sense in cases where only a monomodal or a bimodal size distribution was present (which according to Figures 4 and 7 was the case quite often). This is even visible by eye when looking at Figure 7. Therefore, a more specific approach for the fits should be applied. For example, the authors could apply a trimodal fit only for cases where the fit quality of a bimodal fit decreases (and where a third mode or shoulder is at least recognizable).”

There were several reasons why we always fitted a trimodal size distribution. First, deciding whether to apply a bimodal or a trimodal fit introduces a new ambiguity. The choice between a trimodal and a bimodal fit might be easy to decide in many cases, where it is visible by eye. However, what to do in ambiguous cases? This is a complex problem for which we have not seen a satisfactory answer in the literature. From experience, there are always ill-posed cases in between, where even a subjective decision appears difficult. We agree that a numerical criterion would be needed to decide whether, e.g., the benefits of fitting a trimodal fit would exceed the benefits of the simplicity of just using two modes. Since a trimodal fit will usually always yield a better fit quality, an (arbitrary) threshold in the improvement of the fit quality would be needed to opt between the two choices. Since we analysed a restricted number of average number size distributions, we feel that it is out of scope to make a comprehensive sensitivity analysis for such a threshold criterion.

Second, we certainly obtain a more consistent description of the size distribution data if generally using three modes. If, for instance, the nucleation mode is hardly visible in the size distribution, the corresponding number concentration will be very low. This

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corresponds, in fact, the statement that this mode is “missing”. The main objective of our modal fits was, in fact, a simplified description of the monthly average size distributions given for the different air mass types. We did not aim to provide a conclusive picture on the presence or absence of certain particle modes. We hope that you can accept these arguments.

Nevertheless, we agree that it would certainly be worth to think about the improvement of multi-modal fitting procedures in the future. Such fitting procedures should preferably be applied to the size distributions at their highest time resolution, and not to composite average, such as in our case.

“Section 3.2 / Nucleation mode particles: An analysis of the difference between the CPC number concentration ($N > 10$) and the integrated DMPS number concentration ($N_{20} - N_{470}$) would provide important information on the occurrence of nucleation mode particles.”

We observed indeed new particle formation. But since the analysis of these events was not the scope of our piece of work, we did not put any additional effort to such an examination. In contrast, new particle formation events are analyzed separately and will be presented in a different paper.

“Title: I suggest to be more specific, e.g. “Aerosol number size distributions...””

It is true, the title should be more specific. Therefore we changed it for the revised paper into “Analysis of number size distributions of tropical free troposphere aerosol particles observed at Pico Espejo (4765 m a.s.l.), Venezuela”.

“Page 29159/lines 22 ff: How was the DMPS calibrated?”

The DMPS was calibrated via comparison with another DMPS system in the laboratory in Sweden before it was shipped to the measurement station. After commissioning at the Pico Espejo station flow checks were performed during every service visit to ensure the correct operation of the system. Since this could be a point of interest for readers,

we added this information in Section 2.1 “Observation site and instrumentation”.

“Section 2.2: This section presents data and should be moved to the Result section.”

After reading your comment we agreed that it is indeed better to put this section also into the “Results and discussion” section where it is now Section 3.1.

“Page 29163/line 26: Replace “aerosol densities” with “aerosol number concentrations”.”

This is replaced in the revised paper.

“Page 29164/lines 15 ff: I do not understand/agree with this interpretation. In case of an approximately normal distribution of the values, a lower number of data points will increase the standard deviation but not significantly change the average. Most likely the few remaining data points were just not representative enough.”

We picked up your suggestion and repeated the size distribution fits for monthly averages covering all years 2007-2009. Surprisingly, we confirmed the given structure of extraordinary high accumulation mode diameters in the dry FT aerosol in the months of May, July and October! Therefore, this is now a robust result, and we do not think any more that this might be due to a low amount of data. Meanwhile we have no ready explanation for these changes in mean diameter.

“Figure 6: I do still get the wrong Figure 6 when downloading the manuscript from the ACPD website.”

This was because ACPD obviously did not put a new version of the paper with corrected figures online for discussion. But they received all figures in the right order at the beginning of the submission procedure from us.

“Page 29165/lines 18/19: “Obviously, the dry FT aerosol goes always together with lower specific humidity values compared to the wet FT aerosol”. What do the authors want to conclude from this statement? Isn't it a circular statement?”

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Yes, you are right. The dry FT is of course connected to lower RH values compared to the humid FT, since RH was the criterion for distinguishing between these two FT regimes. Thus this statement is actually not very meaningful and we changed that in the revised paper.

“Page 29166/line 18: Replace “die” by “stop”.”

This is replaced in the revised paper.

“Page 29166/line 19: Add “since particles transported to the site experience...”.”

This is added in the revised paper.

“Summary and conclusions Section / last sentence: This statement on the relevance of the work seems too short to me. I think the authors should be more convincing here, since they indeed have an important and interesting set of data!”

You are right, we should draw more attention to the importance of this research work, since it is somehow a unique set of data. We paid respect to that fact by adding some more sentences about that in the revised paper at the end of the section “Summary and conclusions”.

“Table 1: Please check the significant number of digits.”

We have revised the table with regard to the significant number of digits.

“Table 3: Please remove the decimal digits, they are not significant at all.”

You are right and they are removed in the revised paper.

“Table 4: Please specify the units.”

They are specified in the revised paper.

“Figure 1: In some cases you draw lines between gaps, in other cases you don't. Please check.”

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We revised Figure 1 and for better visualization changed the style of the plot from solid lines into a scatter plot for the daily values and left only the 30-days moving average as a solid line. Now the problem with lines between gaps in some cases and no lines between gaps in other cases is eliminated as well.

“Figure 4: Please specify the year of the measurements.”

The respective period of time is now specified in the revised paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 29153, 2010.

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