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10, C14458–C14466, 2011

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# 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.

### T. Schmeißner et al.

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Received and published: 8 March 2011

#### Author response to Anonymous Referee No. 2

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.

"The title gives an indication of showing more than just the size distributions. The authors could talk about "Physical properties of..." or "Size sistributions of..." in the title."

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It is true, the title could give a wrong impression about the topic of the paper. 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".

"Abstract / line 2: Add FT, "free troposphere (FT)", since FT is used later."

This is added in the revised paper.

"Abstract / line 13: It would be good to mention already here what months are meant with dry and wet seasons. E.g. "dry season (Jan–Mar)"."

Yes, it could avoid some confusion, since there is no general definition of wet and dry seasons. Therefore, we followed your advice and added the respective months here already.

"Abstract / lines 14-15: The same thing for the dry and wet FT, state already here what are the definitions (RH–ranges)."

The RH-ranges are now included in the abstract of the revised paper as suggested by you.

"Intro / Page 29157, line 28: Open "TWI" here since it is mentioned the first time."

You are right, we by mistake introduced the short form of Trade Wind Inversion without any explanation.

"Experimental / Page 29158-29159: Would be interesting to the reader to know what other instruments are available at the site. PSAP is mentioned when discussing the sample flows, something else too?"

Besides the PSAP there is no further aerosol instrumentation on the site, but other devices. As already mentioned there is an automatic weather station. Moreover, 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-

10, C14458–C14466, 2011

> Interactive Comment

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irradiation at 254 nm and a detection limit of 0.4 ppb. We added information about further instruments in the revised paper since it is likely that readers might be interested in that as well.

"Experimental / Page 29159, lines 26-29: How are the DMPS starting from 20nm up and the CPC (> 10nm) data combined? Later on in the fitted size distributions there are many bins between 10 and 20 nm. This could be discussed a bit more."

Unfortunately, there was a misunderstanding and we misspelled the size range of the DMPS. The DMPS measured across a size spectrum of 10nm to 470nm (and later 710nm). The total number concentration of aerosol particles (larger than 10nm) was measured independently with a second CPC. This concentration was then also used to check whether the DMPS was working correctly, by comparing the integral of the DMPS measurements to the total CPC. This information has been corrected in the revised paper and we apologise for causing such a confusion.

"Experimental / Page 29161, the inlet design: It seems that the cloudiness came as a small surprise. The inlet cut-off would be quite an important factor when measuring in–cloud. It seems that an undefined part of the droplets are dried and measured by the system, as also stated by the authors. If the inlet is cutting of some of the droplets, it should be seen in the DMPS data as a sudden decrease in the concentration of the largest sizes. The definition of cloud-cases by just RH is not the most bulletproof way of classification. It's easy to state afterwards, that some other means of detecting a cloud would have been helpful, e.g. a simple webcam. Or different inlet design would have been more useful. Now, with a RH–limit >95% the cloudiness was found to be 40.2%. Would it be much different if 90% was chosen? Some discussion of the possible error due to the RH–selection and inlet selection could be added. Even though I assume it is not changing the results too much."

The cut-off of the whole sampling system is at about 10  $\mu$ m. The limiting factor with regard to the sampling of large particles is in our case not the inlet, but the division of

#### ACPD

10, C14458–C14466, 2011

> Interactive Comment



Printer-friendly Version

Interactive Discussion



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. It is for sure that 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%.

Regarding the technique of using RH as a criterion for classification, we are aware that this is not the securest way. There is for example the problem of patchy clouds which occur at the station quite often and which can be present at RH lower than 95%. But we also tried different RH values as the threshold between cloud and cloud free cases down to 90% RH and the results did not change significantly. Moreover, we have our focus on long-term trends and are averaging over a large amount of data and thus an eventual influence of wrong classified data will be rather small. We mentioned this issue in the revised paper thanks to your and other referee comments.

"Experimental / Page 29162, the fitting of 3 modes: I would like to here more about the mode fitting. Why do you want to always force to provide 3 modes? I would assume that this is not the case in reality all the time. This needs more discussion. The sigma–values are given and fixed. I assume the fitting code has some diameter limits for each mode? Say nucleation mode has to be between 10-35 nm etc. These limits should be presented, and stated that they are also fixed (?)."

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

10, C14458–C14466, 2011

> Interactive Comment



Printer-friendly Version

Interactive Discussion



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 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.

And no, we did not use any fixed diameter ranges for the single modes. Again, the reason is that it is hard to decide which range would be the best choice. An algorithm or numerical criterion would be needed again, as already mentioned for the decision about the number of modes.

Nevertheless, we agree that it would certainly be worth to think about the improvement of multi-modal fitting procedures as well as fixed diameter limits for each mode 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.

"Results: Over all the results section is quite long and hard to read by itself. It leaves many open questions to be remembered until the discussion section. Here, I agree with the comment from Referee No.1, that the readability would improve if results and discussion sections could be combined."

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

## ACPD

10, C14458–C14466, 2011

> Interactive Comment



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Interactive Discussion



number size distributions for the different FT regimes and seasons are discussed immediately.

"Results: The data period used for mode fitting remains a bit unclear for the reader. There are more than two years of data, why only one year is used? Nothing is found in the text, but Table 4 and Figure 5 present data only for 2008. Is that also the case for Figure 4? Need to state clearly in the text and figure/table captions what data is used."

Yes, we first focused only on the year 2008, but of cause 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 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. Concrete declarations of the used data are now made in the revised paper as well and Figure 3 and 5 (former Figure 4 and 5) as well as Table 4 show now results from data covering the whole measurement period.

"Results / Page 29163: The seasons used (months), including the transition periods, should be specified in the beginning."

This specification was already done in section 3.1 "Meteorological conditions at Pico Espejo" (former section 2.1, now changed into the first section of the "Results and discussion" part as suggested by Referee No.1).

"Results / Page 29163, last paragraph: the first (dry 2007) and last (dry-to-wet 2009) seasons are not full in data. This should be noted also more clearly when dealing with the results."

10, C14458–C14466, 2011

> Interactive Comment



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Interactive Discussion



You are right with that and we tried to accentuate this fact more strongly in our discussion.

"Results / Page 29166, the mode concentrations: Some more discussion could be added about how much of the increase/decrease of a certain mode concentration and mean diameter is due to particle growth to the other mode sizes (nucl–ait and ait–accu). Also new particle formation would affect the mode concentrations and diameters. Or is the aerosol transport from lower altitudes still the main driving force?"

As can be grasped, for example, from Figure 7 (old; new numbering: Figure 6), we see rather limited effects of aerosol dynamical growth. In Figure 7c we can see a gradual growth of the Aitken mode, starting between 30 and 40 nm in the evening, and terminating around 40-50 nm at midnight. This could well be the result of particle growth by condensation. The lognormal fitting tracks that growth (Figure 7c) and would thus not convey number concentration, e.g., in to the accumulation mode but keep that concentration in the Aitken mode. Overall, the indications of aerosol dynamical processes visible from the data appear rather minor compared to other observations, such as the particle formation events observed in the boundary layer world-wide (Kulmala et al.: Formation and growth rates of ultrafine atmospheric particles: A review of observations. J. Aerosol Sci. 35: 143-176, 2004, and many others ever since).

Our impression is that the size distribution observations at Pico Espejo are governed by advection, which yields to the visible dramatic changes in the size distributions at given times while the changes of the aerosol within each of those air mass type remains rather limited.

Due to the nature of our single-point measurement, any conclusions about past aerosol dynamical processes in each of those air masses would necessarily remain speculative. This is, in our eyes, a limiting edge of this study.

"Discussion: Not much is mentioned about new particle formation (NPF). Was FT-NPF observed and, if yes, how often and how strong? Or was NPF always occurring at

10, C14458–C14466, 2011

> Interactive Comment



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Interactive Discussion



lower altitudes and transported to the site? Some indication of NPF can be seen in Figure 7."

New particle formation was indeed observed, but there is no deep going analysis about it in the paper since this will be scope of another piece of work.

"Discussion / Page 29171, lines 12-14: Having no major difference in dry and wet FT for nucleation and Aitken modes indicates that there are similar (equally strong) sources for nucleation mode particles in the FT than in the air masses below, right? Or is this an indication of the "forced" nucleation mode in the fitting? In case of a stable Aitken mode, it would be possible to force a stable nucleation mode from the low end of Aitken mode."

Sorry, we do not fully understand the entity of your questions, since so many aspects are interwoven. We will nevertheless try an answer.

First, we agree that the presence of the Aitken mode both, in the FT and the BL, indicates sources of this mode in both compartments. It is likely that the nucleation mode is generally the main feeder for the Aitken mode, especially in the FT. Meanwhile, the deposition or loss rates of Aitken mode particles are much lower than for the nucleation mode. This concerns, e.g., below-cloud scavenging; also, nucleation mode particles are much more prone to self-coagulation and coagulation with bigger particles. The nucleation mode is thus expected to be more variable in time than the Aitken mode. This is confirmed by our observations.

About the lognormal fitting: We suspect that you wonder about the continuous presence of a nucleation mode in our lognormal fits. As now stated in the manuscript, the nucleation mode was always added to the fit for reasons of consistency (trimodal fit always). Therefore, the nucleation mode is present in the data all the time, but most of the time with a very small number concentration. In those situations you could well say that its presence is "virtual" because the nucleation mode represents most of the time just the left shoulder of the Aitken mode. We nevertheless preferred to keep the

# **ACPD**

10, C14458–C14466, 2011

> Interactive Comment



Printer-friendly Version

Interactive Discussion



nucleation mode all the time for reasons of consistency.

*"Figures: Figure 2 and 3 show basically the same results. I feel Figure 3 is more informative and Figure 2 could be omitted."* 

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.

"At the end it would be interesting to know if the very nice measurement site is still running or was this a campaign for two years. Related to other possible measurements being onsite, it would be interesting to hear what we can expect next from the authors. What is the next step with the data set?"

Measurements are on hold at the moment since the cable car is out of order. As soon as repair of the cable car is done the measurements are supposed to be carried out again. Concerning other work, an analysis of observed new particle formation is ongoing.

"Abstract / line 2: "Mai", correct to "May" throughout the manuscript."

This is corrected in the revised paper.

"Abstract / lines 7-8: "Differential Mobility Particle Sizer (DMPS system)", correct to "Differential Mobility Particle Sizer (DMPS) system"."

This is corrected in the revised paper.

"Intro / Page 29157, line 12: "Headley", correct to "Hadley" throughout the manuscript."

This is corrected in the revised paper.

"Figures: The text in Figure 7 is very small. The font could be increased."

The font is increased in the version for the revised paper.

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

# ACPD

10, C14458–C14466, 2011

> Interactive Comment

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Interactive Discussion

