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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long term in-situ observations of biomass burning aerosol at a high altitude station in Venezuela – sources, impacts and inter annual variability" *by* T. Hamburger et al.

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We thank the reviewer for his detailed and valuable comments on the manuscript. The reviewer's comments are in regular type and our responses are outlined in italic type.

General overview

This manuscript presents a 2,5 year set of aerosol measurement data from a high-altitude site near the savanna region of northern South America. The measured aerosol parameters are particle number size distribution (12-500 nm), size-dependent

non-volatile particle number concentration (300 C) and aerosol absorption coefficient. No other long term measurement campaigns have been published from this area, but part of this measurement data has been published in a paper by Schmeissner et al.,2011. This new manuscript looks at the data from a different point of view, and does not conflict or take credit from the earlier paper. The results presented in this manuscript are new and important for the scientific community.

The instrumentation and data analysis is generally well described and valid. The authors clearly state the reasons to use or exclude certain parts of the data. The two-altitude trajectory analysis used in the manuscript is a nice approach to cope with the problems trajectories have in complex terrain. The main results are stated clearly and discussed well. Also earlier work is cited and credited properly.

The overall structure of the paper is clear, but as one looks closer to the individual chapters, the focus of the text often jumps from one topic to another and then back. This, combined with complex sentence structures, makes the manuscript unnecessarily difficult to follow. Streamlining the text would make it a lot more reader friendly.

The number of tables and figures is appropriate for the text, and there is no need to add or remove any tables or figures. More precise comments are given in the end of this referee comment under section "Tables and figures" In general this is a good manuscript presenting valuable data and results. I suggest this manuscript to be accepted with modifications stated below:

Major comments

Data and analysis:

The authors have measured size-dependent number concentration of particles, but report particle volume concentration throughout the text. The manuscript should state how the number concentration data was converted to volume concentration and what assumptions were made.

We added the following sentences to Section 2.3 Data analysis:

The particle volume concentration was calculated from the particle number size distributions assuming spherical particles. One has to keep in mind that the number size distribution was inverted using the mobility diameter (volume equivalent diameter in case of spherical particles). Depending on the real morphology of the observed particles, the assumption of a spherical shape might slightly overestimate the volume concentration. In the atmosphere, particles such as soot agglomerates are relatively compact if they have been in contact with water and the shape factor might be only slightly larger. One can assume that most biomass burning plumes reaching the observation site have been aged for several days. Additionally, condensation of secondary particulate matter on soot particles leads to an even more compact form (Zhang et al., 2008; Fu et al., 2012) which reduces the error created by the assumption of spherical particles.

The reported measurable particle size range is (10-500nm) different from Schmeissner et al., 2011 (10-470nm), even though the instrument and data are the same.

Thank you for this comment to improve the quality of the manuscript. The size range in the present manuscript was an oversimplified rounded value. It is changed to the precise value of 470 nm throughout the manuscript.

Also the seasons are defined differently. It would be good to discuss the differences and add some more information of the ambient particle number concentration and size distribution in the text, even though they are reported in the Schmeissner et al., 2011 paper.

In the recent paper we define the seasons for the Venezuelan savannah after Morales et al. (1990) and Rondón and Sanhueza (1990). The definition depicts the typical separation of the tropical meteorological year into two seasons – wet and dry – and is

based on "local", citable publications. However, there is no sharp change from the dry to the wet season and the transition period may change slightly from year to year. This can be seen in the time series in Figures 2 and 3 in the present manuscript. Thus, a subdivision into inter-seasonal periods as done by Schmeissner et al. (2011) is a valid procedure for a more detailed analysis of inner-annual variations. It is more suitable for a detailed analysis of specific process related particle properties (e.g. shape of the aerosol size distribution in contrast to the total aerosol volume). The classical division into two seasons proved to be sufficient in the present analysis which serves as an overview of the presented particle properties.

Text streamlining throughout the manuscript, especially following chapters:

3.2 Seasonal cycle: Please re-structure the chapter so that you first describe the conditions for dry season and then for wet season, and the same for LFT and BL. In the current form the chapter is very difficult to follow.

Thank you for pointing this out. We restructured the chapter for more readability as follows:

Table 1 summarises the mean values of each season observed within the LFT and the BL.

The relative humidity (RH) shows a high variability in the LFT for both, dry and wet season with a standard deviation between 25–27 % (see also time series in Fig. 2). However, RH increases from 45 % to 67 % from the dry to the wet season. The temperature remains constant at $t \sim 0$ C. The average particle number and volume concentrations within the LFT during the wet season can be considered as mean background values which are rather stable throughout the year. The particle volume V reaches an average of 0.19 $\mu\mathrm{m}^3\,\mathrm{cm}^{-3}$ and the average absorption coefficient b_{abs} is 0.15 Mm^{-1}.

The relative humidity and ambient temperature remain constant throughout the

seasons for BL air masses (RH = 82–86 %, $t \sim 2$ C). The meteorological seasonal cycle has a large impact on all observed aerosol parameters within the BL. The particle volume V reaches an average of 0.49 $\mu m^3\,cm^{-3}$ in the wet and 1.4 $\mu m^3\,cm^{-3}$ during the dry season. The absorption coefficient b_{abs} increases from 0.32 Mm^{-1} to 0.91 Mm^{-1} (factor 2.8) from the wet to the dry season. However, both parameter show a standard deviation of $\sim$ 100 %. Recent studies showed an increase of b_{abs} from the wet to the dry season from 0.5 Mm^{-1} to 2.8 Mm^{-1} (factor 5.6) in the Amazon rain forest at 110 m a.s.l. (Rizzo et al., 2012). This is twice the increase of b_{abs} compared to our observations of biomass burning in the savannah. The trajectory analysis shows the highest average number of fire events since the last precipitation event of 3 for the dry BL. This is consistent with the increased particle load during the biomass burning season.

5.2. 12 yr of satellite observations: This chapter first states the meteorological conditions as the main reason for inter annual variability in aerosol conditions. Then it describes the regulations and then back to meteorology. I suggest discussing the meteorology first and then the regulations. Also in the meteorology part it is unclear for some sentences whether they describe El Nino or La Nina conditions.

This chapter starts with a description of the observed decrease of biomass burning from 2007 to 2009.

It continues with an overview of foregoing studies which observed similar trends in other South American countries. This paragraph also states the explanations of the foregoing studies for the observed trends.

The following paragraph gives information on Venezuelan regulations and describes that the effect of the regulations cannot be seen yet in the observations which were analysed in the present manuscript.

The last part of the chapter describes the observed influence of ENSO on the biomass

burning activity.

We consider this structure as useful and would like to retain this structure.

However we changed the text as follows to avoid confusion about El Nino or La Nina conditions. We excluded lines 14–18 on page 13099 (Gianni et al. ... entire basin) as the opposing effects of El Niño on the precipitation in different areas of the Caribbean seem to distract the reader. Additionally only the information on the northern part of South America is required. However we retain the citation and include it in the previous sentence. We changed the following sentences:

We added information on sea surface temperature (SST) anomalies in the Pacific region to the time series in Fig. (8) to indicate warm and dry episodes (El Niño) and cold and wet episodes (La Niña).

[...]

Interestingly, a decrease in burned area can be seen for consecutive warm and dry El Niño episodes. After the intense biomass burning season in 2003, the burned area decreases from 2004 to 2005 – all three years featured El Niño conditions. This indicates that a lack of burning fuel and less demand on agricultural biomass burning can also result in less burned area. On the other hand, all intense biomass burning seasons feature El Niño conditions, which follow years of cold and wet La Niña conditions (e.g. 2007 and 2010). I.e. less biomass burning but more intense regrowth of grass and scrubs was possible before the warm and dry El Niño period began. This means that more burning fuel was available in a biomass burning season during a warm and dry El Niño episode, if it followed a cold and wet La Niña episode.

Minor and technical comments

It would be nice to point out in the manuscript that the number-size-distribution of the aerosols is already examined in Schmeissner et al., 2011 and is therefore not included

in this manuscript.

We included the following to chapter 2.2 Instrumentation:

The aerosol instrumentation was provided by the Department of Applied Environmental Science (ITM), Stockholm University (see Schmeissner et al., 2011, for details).

[...]

The analysis of the observed ambient aerosol number concentrations at the Pico Espejo was realised by Schmeissner et al. (2011) and can be found in the respective publication.

P13082 L10: There is a lot of other recent research besides Myhre et al., 2009 aiming to reduce the uncertainty of the climate effects of aerosols. Since you can not list them all, I suggest using form (eg. Mygre et al., 2009), with possibly some other sources mentioned as well.

This is of course true. We added the respective references:

Recent and ongoing work helps to reduce these uncertainties (e.g. Forster et al., 2007; Myhre, 2009; Shindell et al., 2009, and references therein).

P13082 L11-12: "Within . . . Troposphere" I suggest moving this sentence in the last chapter of the introduction, as that is where you describe other things about your own work.

The purpose of this sentence is to motivate the focus on biomass burning aerosol in the following paragraphs of the introduction. Therefore we prefer to keep the sentence on place - even though the sentence contains some information on the content of the paper which is again described in the last chapter of the introduction.

P13082 L21-22: "Unlike . . . sunlight." This sentence is not needed, as the same thing is said in the previous paragraph.

We removed the sentence.

P13084 L12 "It . . ." Please replace word "It" by "The station" to make it more clear that you are not talking about ZFK or KIT anymore.

Thank you for pointing that out. We replaced it.

P13084 L12: Is the eastern branch of the Andes is roughly like a single ridge or more complicated topography in this area.

This depends of course on the scale at which we are looking on the mountain range. On a large scale, one can look at it as a single ridge comparable to the central ridge of the Alps. On a smaller scale, the valleys feature a rather complicated structure. We added the following information to chapter 2.1 Measurement site:

The mountain is part of the North-eastern branch of the Andes. The mountain range has a width of $\sim$100 km at the location of the observation site.

P13084 L13-18: I suggest to include the altitudes of the cities in the text to make it clear that the station is at clearly higher altitude.

We included the altitudes of the cities in the text.

P13085 L21: "The residual . . . aerosol." Even though this sentence is probably true, there is nothing in your measurements that can prove it. You can only assume that they are primary particles.

We changed the sentence to:

The residual particles include primary aerosol as well as very low-volatile secondary organic compounds (Wehner et al., 2005; Häkkinen et al., 2012). The primary...

P13085 L26-29: "Particles . . . plumes (hours)." Is this also from Janhäll et al., 2010?

Yes - we moved the citation to the end of this sentence.

P13086-13087: I suggest streamlining the text such that you first report what data you remove from both DMPS and PSAP (cloud periods), then go through one instrument at the time for further exclusions and then tell how much data is left for each instrument. It could also be good to estimate how much error is caused by the exclusions. Finally in the end keep the description of the LFT / BL division.

We moved the paragraph which explains the detection and removal of in-cloud data to the beginning of Section 2.3 Data analysis. It is followed by the analysis of DMPS data and PSAP data. The section ends with the number of total excluded data and the division into LFT/BL.

P13089-13090 L29-2: "We were . . . boundary layer" This sentence remains unclear. Please consider rephrasing.

The sentence seems to distract the reader and the information is not necessarily required here as it is part of the result section. We decided to remove the sentence to focus on the description of the method.

P13090 L13-14: "The ITCZ . . . Hadley Cell." I suggest removing this sentence, since it is not needed for the analysis, but distracts the reader.

The sentence is removed.

P13092 L12-13: "The average RH . . . observation period." Please rephrase.

We rephrased the sentence as follows:

In the LFT the average RH increases from the dry to the wet season from 30–80 %. However, it features a high daily variability throughout the observation period.

P13093 L9-13: It would be good to point out that these numbers include both LFT and BL values.

We included:

The whole dataset combing BL and LFT data was used for the calculation of the diurnal cycles. However,...

P13094 L10-12: "In addition, . . . possible sources." In the introduction you state that Rissler et al., 2006 states that 30% of global biomass burning activities take place in entire South America. Here you give the same value and reference for the Amazon basin. Please correct the one that is wrong.

Thank you for spotting this mistake. The 30 % take place in the entire South America. However, the value is a re-re-citation to a non-accessible source. So we delete the citation in the introduction as additional citations already emphasise the importance of South American biomass burning on the global BC. In addition we add the following to P13094 L10-12:

Further, the Amazon region may serve as a possible source region of biomass burning aerosol. Approximately 40 % of the total particulate mass which is released by deforestation activities in the Tropics originates from biomass burning in the Amazon region (Longo et al., 2009).

P13094 L18-21: In the figures figure 5a is without precipitation and figure 5b with precipitation taken into account, here the opposite way. Please correct.

Thank you for spotting this mistake. We corrected it in the text.

P13095 L12: ". . .precipitation event." Please add a reference to figure 6.

Reference is added.

P13096 L15-16: "But, not only . . . important." This sentence is unclear. What do you mean by individual properties of the plume? (Is that each property of the plume alone or properties of individual plumes?) P

To avoid redundant information and to clarify the paragraph we drop lines 13-16 on page 13096 (The resulting ... important.)

P13096 L17-21: "Sakeada . . . radiative effect." Please check the signs of the effects, and consider using warming and cooling effect to avoid misinterpretation.

We rephrased the paragraph as follows to emphasize the description of a top of the atmosphere effect:

At the top of the atmosphere semi direct radiative effects can differ in sign depending on the location of the biomass burning plumes and their underlying surface, i.e. if they are located above land (positive semi direct radiative effect) or ocean (negative semi direct radiative effect) (Sakaeda et al., 2011).

In addition we changed lines 21-23 on page 13096 to clarify the content:

Plumes containing soot particles change the local atmospheric stability due to the ability to heat the ambient air by absorbing the solar radiation. Hence, elevated biomass burning layers have the potential of additional lifting, increasing persistence

and thus longer lifetimes in the free troposphere (Boers et al., 2010).

P13097 L12: "The transport . . . in Fig. 7a and b." What do you mean by dashed clouds?

We changed the expression to:

... which are indicated by dashed lines...

P13097 L22: Please use 12 years instead of 12 yr in the section header.

Changed.

P13098 L10-11: Please include a reference or more description about the tri-national policy shift.

We added the reference Brown et al., 2006

P13098 L23-26: "Those brigades . . . across the country." Explaining the work of the communal fire brigades is clearly outside the scope of this manuscript. Please exclude this sentence.

The sentence is excluded.

P13098 L27-30: "So far, . . . legal regulations" Here I can not follow your reasoning. How do the lack of trend indicate success in implementing legal regulations?

This is a misinterpretation. We see no trend, i.e. there is no indicator of a successful implementation of regulations. To clarify we rephrased the sentence as follows:

So far, the time series of the burned area per month does not show a continuous trend

within the years 2000 to 2013. Thus, the analysis does not show an indication for a successful implementation of the legal regulations, yet.

P13100 L24-25: "No evidence of long range transport . . ." Since the Amazonas plumes are described in previous sentences, I assume that this means transport from Africa. It would be more clear to rephrase the sentence into "No evidence of long range transport from Africa . . ."

We changed the sentence according to the suggestion.

Tables and figures

Figure 1: Extending the map a bit more to the west would be nice. Now Pico Espejo is close to the edge of the map even though it is the downwind edge.

Thank you for the suggestion. However, we prefer to keep the map boundaries to set focus on possible source regions upwind the observation site.

Figure 2: Adding time series of N would be nice. Also using smaller maxima for the Y-axis of V and b_{abs} would improve the readability.

We added the time series of N and adjusted the Y-axes (see supplement).

Figure 3: Same suggestions as to Figure 2.

Same changes as to Figure 2.

Figure 4: Similar suggestions than to Figures 2 and 3.

Similar changes as to Figures 2 and 3.

For consistency we applied the same changes to Figure 6.

We added/changed the following text to consider the added ambient number concentrations in the figures:

P13092 l28:

A similar trend can be observed for the ambient particle number concentrations N (1500/1200/750 cm^{-3}) and the refractory particle number concentrations $N_{300\,\mathrm{C}}$ (1300/720/470 cm^{-3}).

P13093 l14-18 deleted for text streamlining

P13093 l19 (text added):

During the dry season the ambient particle number concentration reaches its plateau of maximum concentrations between 12:00–17:00 LT. The ambient particle volume concentration and the refractory particle number concentration continuously increase until they reach their maxima between 14:00–16:00 LT. The early increase in ambient particle number requires additional sources next to the advection of existing particulate matter. The results of Schmeissner et al. (2011) indicate the possible presence of new particle formation. New particle formation was also observed during prior studies at high altitude observation sites and aircraft studies within different climatic regions (e.g. Weingartner et al., 1999; Venzac et al., 2008; Kivekäs et al., 2009; Hallar et al., 2011). However, a detailed analysis of new particle formation observed at the Pico Espejo would be beyond the scope of this paper and has to be accomplished in further work.

The rather high mean values of babs in the early afternoon hours compared to its median values result from single events. babs reaches up to 8–10 Mm^{-1} during those episodes of high absorption.

P13095 l25 (text added):

The average ambient particle number concentration increases from 500 cm^{-3} at 0–5 fire events to 1700 cm^{-3} at 45–50 fire events. The increase in number can most frequently be associated with new particle formation, which must not necessarily be linked to biomass burning. However, the trajectory analysis indicates that biomass burning contributes to a significant amount to the increase in number concentration. Secondary biomass burning products such as sulphate or organic species are released by fires next to primary particles (Reid et al., 2005; Pratt et al., 2011) and might contribute to an increase in particle number concentration and certainly in particle volume concentration.

Figure 5: Concentration this figure to only South America would increase the readability.

We changed the boundaries of the maps to get a closer look on the source areas. However, we still include a small part of West Africa.

Figure 8: Seasons in plural in the figure caption.

Thank you for spotting this. We changed it.

References

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C5705/2013/acpd-13-C5705-2013-supplement.pdf

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