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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.

T. Hamburger et al.

thomas.hamburger@nilu.no

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

This paper presents long term observations of South American aerosols measured at Pico Espejo Atmospheric Research Station located at 4765 m asl. The measured aerosol parameters are particle number size distribution, size-dependent non-volatile

particle number concentration and aerosol absorption coefficient. Data is clearly presented, and the instrumentation is adequately described.

Major Comments:

1. Several papers have shown that New Particle Formation (NPF) is prevalent at high elevations, and strongly associated with the diurnal cycle. Here are examples:

Venzac, H., Sellegri, K., Laj, P., Villani, P., Bonasoni, P., Marinoni, A., Cristofanelli, P., Calzolari, F., Fuzzi, S., Decesari, S., Facchini, M.-C., Vuillermoz, E., and Verza, G. P.: High, Frequency New Particle Formation in the Himalayas, PNAS, 105(41), 15666–15671, 2008.

Hallar, A.G., D. H. Lowenthal, G. Chirokova. C. Wiedinmyer, R.D. Borys, 2011: Persistent Daily New Particle Formation at a Mountain-Top Location, Atmospheric Environment, doi:10.1016/j.atmosenv.2011.04.044.

This work does not consider the potential influence of NPF on the aerosol concentration. It appears that diurnal change in aerosol concentration was connected directly to the BL, yet NPF may play a major role here. Were the size distributions plotted as contours to look for "banana plots"?

From Schmeissner et al., 2011, Figure 6a, it appears that NPF may be occurring at this site. This should be explored further, as biomass burning may not be the sole aerosol source.

Thank you for the comment on this very interesting topic. The authors are aware of the phenomena of frequent observations of new particle formation (NPF) at high altitude sites. Further detailed analysis of the observed size distributions revealed that NPF also occurs frequently at the Pico Espejo. The detection and separation of time series data of NPF events or NPF indications required a more complex methodology and the results are prepared for a separate manuscript. NPF were not mentioned in the present manuscript so far as the content focusses on the effect of biomass burning on primary *particles and the total particulate burden in the observed size range. Adding an in-depth analysis of NPF would bloat the scope of the present manuscript and distract from its original purpose. However, we added the aerosol number concentration to Figures 2, 3, 4 and 6. Thus, the following content (including the mentioning of NPF) was added to the text:*

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 1 700 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.

2. The statement in section 2.2 on line 3 is an oversimplification. Several papers have observed dust at high altitude stations in remote locations. Here are a few examples:

Holben, B.N. D. et al., 2001, An emerging ground based aerosol climatology: Aerosol Optical Depth from AERONET, J. Geophys. Res., 106, D11, 12,067-12097.

VanCuren, R.A., Cliff, S.S., Perry, K.D., Jimenez-Cruz, M., 2005: Asian continental aerosol persistence above the marine boundary layer over the eastern North Pacific: Continuous aerosol measurements from Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2). J. Geophys. Res. 110, D09S90 .

Fischer et al., (2009) A decade of dust: Asian dust and springtime aerosol load in the U.S. Pacific Northwest, Geophysical Res. Letter, 36.

VanCuren, R. A., and T. A. Cahill (2002), Asian aerosols in North America: Frequency and concentration of fine dust, J. Geophys. Res., 107(D24), 4804, doi:10.1029/2002JD002204.

Hallar, A.G., G. Chirokova, I.B. McCubbin, T.H. Painter, C. Wiedinmyer, C. Dodson, 2011: Atmospheric Bioaerosols Transported Via Dust Storms in Western United States, Geophysical Res. Letters, 38, L17801, doi:10.1029/2011GL048166.

We removed the oversimplification and added some of the mentioned references:

However, the appearance of larger primary particles in the super-micrometre diameter range such as sea salt or mineral dust can-not be completely neglected due to long range transport (e.g. VanCuren et al., 2005; Ben-Ami et al., 2010; Hallar et al., 2011).

3. More detail is required regarding the cloud detection algorithm.

We added the following details to explain the cloud detection algorithm:

The webcam used for cloud detection was facing towards the South-east. The horizon split the webcam images at approximately 40 % from the bottom. The upper 60 % of the images showed the sky. The Python Imaging Library (PIL) was used to detect the relative fraction of blue, grey, black or other pixels. Clear sky was defined if the upper 50 % of the picture consisted of more than 75 % blue pixels and the total amount of blue pixels was less than 60 % to avoid incorrect imagery. If grey could be assigned to more than 90 % of the total pixels the image was defined as in-cloud image. Night-time was defined if more than 50 % of the total pixels were black. The assigned in-cloud sequences had a median relative humidity of RH=97 %. When no suitable webcam image was available - or during the night - all observations with RH> 95 % were marked as in-cloud sequences and the respective data were excluded for the subsequent analysis.

4. Using local daylight time to define the lower free troposphere from the boundary layer is an over simplification. This is highlighted in section 3.3 and Figure 2, where you state an observation of 80% RH in the lower free troposphere. This definition then leads to the unsupported conclusion that soot is mixed into the lower free troposphere.

Thank for this interesting comment. We plotted the time series of relative humidity, particle volume concentration, refractory particle number concentration, and absorp-

tion coefficient in Figure 1 (see supplement). The time series shows data selected for the LFT by local time and/or RH as described in the manuscript (grey). In addition, observations at relative humidity between 0-70 % (green) and 0-50 % (red) are presented. Observations of BL air masses can already be excluded at RH<70 % as the average relative humidity remains in the BL throughout the year above RH$\sim$75 % (see Figure 3 in the manuscript). However, no substantial difference in the particle concentrations can be observed compared to the time series of LFT data (as selected in the manuscript). A decrease of particle concentrations can be observed if the time series data is limited to RH<50 % when dry subsiding air masses from the upper free troposphere reach the observation site. The advantage of using local time and/or RH as an indicator of LFT air masses is higher data coverage. The relative humidity might excess a RH threshold of e.g. RH=70 % especially during the wet season when convection upwind the observation site increases the RH also in the LFT (see Schmeissner et al., 2011).

Minor Comments:

1. Throughout the introduction, there are many citation incorrectly listed as the sole source, for example the first sentence of the manuscript. Please add e.g. before these.

We added "e.g." before citations to general information.

2. More detail should be presented regarding the transmission efficiency of the aerosol inlet. What is the 50% size cut off?

We added the following information to p13085 l10:

The aerosol inlet has a transmission efficiency close to 100 % for submicron aerosol. The 50 % cut off diameter is $D_{50\,\%}$ $\sim$5–7μm.

References

Ben-Ami, Y. et al., Transport of North African dust from the Bodélé depression to the Amazon Basin: a case study Atmospheric Chemistry and Physics, 2010, 10, 7533-7544

Hallar, A. G. et al., Atmospheric bioaerosols transported via dust storms in the western United States Geophysical Research Letters, 2011, 38, L17801

Hallar, A. G. et al., Persistent daily new particle formation at a mountain-top location Atmospheric Environment, 2011, 45, 4111 - 4115

Kivekäs, N. et al., Long term particle size distribution measurements at Mount Waliguan, a high-altitude site in inland China Atmospheric Chemistry and Physics, 2009, 9, 5461-5474

Pratt, K. A. et al., Flight-based chemical characterization of biomass burning aerosols within two prescribed burn smoke plumes Atmospheric Chemistry and Physics, 2011, 11, 12549-12565

Reid, J. S. et al., A review of biomass burning emissions part II: intensive physical properties of biomass burning particles Atmospheric Chemistry and Physics, 2005, 5, 799-825

Schmeissner, T. et al., Analysis of number size distributions of tropical free tropospheric aerosol particles observed at Pico Espejo (4765 m a.s.l.), Venezuela

Atmospheric Chemistry and Physics, 2011, 11, 3319-3332

VanCuren, R. A. et al., Asian continental aerosol persistence above the marine boundary layer over the eastern North Pacific: Continuous aerosol measurements from Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) Journal of Geophysical Research: Atmospheres, 2005, 110, D09S90

Venzac, H. et al., High frequency new particle formation in the Himalayas Proceedings of the National Academy of Sciences, 2008, 105, 15666-15671

Weingartner, E. et al., Seasonal and diurnal variation of aerosol size distributions (10 750 nm) at a high-alpine site (Jungfraujoch 3580 m asl) Journal of Geophysical Research: Atmospheres, 1999, 104, 26809-26820

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C5723/2013/acpd-13-C5723-2013-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 13079, 2013.