

## ***Interactive comment on “A comparison of dry and wet season aerosol number fluxes over the Amazon rain forest” by L. Ahlm et al.***

**Anonymous Referee #1**

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*In this manuscript vertical number fluxes of aerosol particles and CO<sub>2</sub> are presented from an Amazon rain forest during the (The Large Scale Biosphere Atmosphere Experiment in Amazonia) experiment. The measurements were obtained during the wet and dry season in 2008. The aim of the work was to quantify the forest dry deposition sink for both wet (clean) and dry (more polluted) seasons. The study also investigated whether there was any particle emission observed from the surface and showed that under certain circumstances this could dominate over dry deposition sink.*

The data is presented in a clear and methodical manner and interpretation and description of diurnal patterns in CO<sub>2</sub>, energy budgets and particle fluxes appear to be correct. Some of the conclusions associated with particle fluxes, e.g. emission, are tentative, since the data sets appear to be limited by a lack of information on particle

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size, a critical missing component. Despite this the general conclusions appear sound.

The abstract should include if possible some brief information on the size of the particles measured to which the dry deposition velocities refer to. As  $v_d$  can vary hugely as a function of size between the ultrafine and accumulation modes. It is important that some information on this metric is included, whether based on a number median size with a geometric standard deviation of the size distribution and/or a volume median metric. Without this information this information on the relationship presented is not readily usable or comparable to other studies. If this information is not available then the authors should not be afraid to state this (it is a difficult environment to work in) and further reference to other studies used to reinforce the conclusions might help.

The quoted relationships should also include as a matter of course the uncertainty associated with the regressions used to derive them.

The conclusion as to the small difference between the relationships between wet and dry seasons is “*The reason is probably domination of accumulation mode particles in the Amazon boundary layer, both in the dry and wet season, and low wind speeds in the tropics compared to the midlatitudes.*” This is a reasonable conclusion but I am surprised there is no measurement of number size distribution to support this, particularly as during the dry periods when it is more polluted biomass burning periods one might expect a shift in the size distribution of the aerosol. Can the authors show a typical size distribution to support this? As stated the above sentence from the manuscript can be confusing. Much later in the manuscript we learn that all these conclusions are based on measurements of fluxes using an ultrafine particle counter (sensitive to particles > 10 nm) i.e. we do not know whether the size distributions are dominated by sub 100 nm or accumulation mode particles per se. It should be stated in the title and the abstract as well as much earlier in the manuscript that what is being presented are *likely* fluxes and deposition velocities associated with accumulation mode particles so as to assist the reader and place the measurements in a suitable context, i.e. likely have little to do with PM mass fluxes and will have very small deposition velocities. Again without num-

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ber size distribution measurements to assess this statement it makes it very confusing for the reader.

The counting efficiency of condensation particle counters is known to degrade with time due to contamination, faster or slower depending on ambient PM loadings, and/or water uptake. How was the instrument monitored and calibrated to check for this?

The authors state: line 35 –

“It is possible that these morning upward particle fluxes are associated with emission of natural biogenic particles from the rain forest.” The implication is that biogenic aerosols are being formed within the canopy. Are we to assume from this that these particles represent a different population compared to the ambient “likely” accumulation mode particles which they see as being deposited for the majority of the time? And that these biogenic particles may occupy a different size range? Most primary biogenic particles are “likely” supermicron in size and with very low concentrations, Bioogenic secondary organic aerosol will likely be sub-micron - are the authors suggesting these particles are primary or secondary? Unfortunately without size resolved measurements there is no proof presented in the manuscript to support this statement. The question that seems to be difficult to address is -what are these particles composed of and what size are they?

Further more they go to say – line 37/38

*“Emitted particles may be stored within the canopy during stable conditions at night-time, similarly to CO<sub>2</sub>, and being released from the canopy when conditions become more turbulent in the morning.”*

Again the implication is that these particles are being produced by some mechanism within the canopy. The data as presented is not a *prior* proof of any in-canopy particle production and this needs to be made clear. Does this statement also imply that the production occurs at night-time or that it does occur during the day but any emission

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is masked by deposition and only seen during the breakup of the nocturnal inversion in the morning period. The statement implies that the majority of these particles are produced or “emitted” inside the canopy? This needs to be clarified. If the particles are emitted/produced within the canopy at night and/or during the day and are not seen above the canopy at this time due to low turbulence levels, why should this not be a continuous process that results in a sustained flux but is only seen when turbulence levels are sufficient during the day?

Having said this, Figures 5 and 6 contradict this conclusion as it is obvious that “emission” is indeed occurring during the night time period to levels almost as high as those seen first thing in the morning? The “significance” of these flux deviations needs to be quantified with respect to the observed apparent emissions both first thing in the morning and at night, see e.g. Pryor et al. 2007 who suggest using the following to identify whether a flux is significant - “The assessment of a ‘significant’ flux is rather subjective. Here we use a threshold,  $|F| - \delta F > 0$ , noting that for a Gaussian distribution, 68% of data values lie within  $\pm 1\sigma$  of the mean.”

It is not clear whether the flux data were quality controlled using standard stationarity criteria (see Foken 2007). Under conditions of low turbulence, small positive heat fluxes (at night) and low winds (convective conditions) during the day, a significant fraction of the fluxes may fail the usual data quality control criteria. Have these criteria been applied to filter these periods from the data set?

It is possible that false correlations between friction speed and fluxes can occur due to trends in the data particularly when the dynamic range of the variable (in this case wind speed/friction velocity) is so small with few data points at the extreme of the range. The graphs showing particle deposition velocity as function of  $u^*$  show significantly increasing errors with increasing  $u^*$ . How significant are the trends in these graphs? The uncertainty in the trends should be quoted in the formulations for each season.

Could the authors provide a plot of particle number flux versus concentration, as dis-

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cussed in e.g. Flanagan et al. (2005) and derive a relationship between the two, to see if this is consistent with a sudden change from an emitting “source” to a sink within their micrometeorological flux footprint and to show the scatter in their data. This is only a suggestion.

I would suggest that the deposition velocity data can be desegregated into both surface as well as aerodynamic transport components quite easily using the usual resistance analogy approach and if plotted as a diurnal median for both wet and dry periods might elucidate the actual surface deposition process better. Again this is just a suggestion.

Can the authors suggest a formation mechanism for their implied production of “likely” sub-micron/ultrafine particles that would be consistent with current understanding of new particle nucleation or bio-degradation of plants by bacterial decay? Particles that are deposited to the canopy can presumably be subsequently removed by detrainment/venting through action of enhanced turbulence as the daytime convective boundary layer starts to form breaking through the nocturnal inversion.

Figure 5b. This is a nice plot of a diurnal deposition flux for ultrafine particles and looks to be very consistent with current knowledge of this process. It also lends weight to the formulation for  $v_d$  versus  $u_*$ . I do not think Figure 5a, adds anything and could be removed or the percentiles added as shaded regions in 5b. The small emission between 07:00 and 08:00 is consistent with venting of the canopy by increased turbulence as canopy heating proceeds as concluded by the authors. However, compared to the excursions at night (one might have expected the errors here to be larger due to the much smaller turbulence levels at this time?) they are not significantly different (at least no significance comparison is provided) and without more detailed measurements it is really impossible to state that these particles are due to any biogenic production mechanism in the canopy – although it is suggestive.

Line 534. “In general, deposition velocities are low here compared to studies over boreal forests (Ruijgrok et al., 1997; Buzorius et al., 2000; Gaman et al., 2004; Pryor

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et al., 2007). Dominance of accumulation mode particles in Amazon boundary layer, both in the dry and wet season, is one explanation for these low values on  $v_d$ .”

This statement is probably correct but needs to be strengthened by more detailed reference to measurements in the literature particularly given that no particle size information has been provided.

For example – in Figure 1, Pryor et al. (2007) as cited here, measurements above forests by Pryor et al. and by Gronholm et al. show significant differences in  $v_d$  but typically range from 2.5 – 4.5 mm s<sup>-1</sup> (for particle diameters ranging from 0.1 to 0.025  $\mu\text{m}$ ) in the former to 6 – 15 mm s<sup>-1</sup> (for particle diameters ranging from 0.1 to 0.02  $\mu\text{m}$ ) for the latter. In this manuscript (line 525) we are told that the deposition velocities are such that. . . “In both seasons,  $v_d$  peaks at approximately 1 mm s<sup>-1</sup> in the middle of the day or early afternoon.” So yes the statement is correct, the velocities are “significantly” smaller. However, although the measurements cited in Pryor et al. show considerable scatter the typical minimum reported for previous forest studies is approximately 2 mm s<sup>-1</sup> in the accumulation mode size range  $> 0.1 \mu\text{m}$ . Which is still “significantly” larger than presented here. Secondly it is not clear from the Pryor et al citation whether average or median values of  $v_d$  have been quoted.

## 1 Minor typographical errors

Figure 1 is rather “fuzzy” and needs to be improved.

Line 140. “using a . . .”

Line 247. Actually the equation governing the uncertainty in aerosol fluxes associated with particle counting instruments was first stated by Fairall (1984), the citation and equation described here is a re-statement of this original work. Fairall should also be quoted.

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Line 304. "...during the afternoon."

Line 305: "As was discussed in Ahlm et al. (2009), much information of the diurnal cycle of the boundary layer can be revealed by investigating the diurnal cycle of water vapor concentration (Fig. 2e). "

I suggest this be re-worded to make it flow easier – "As discussed by Ahlm et al. (2009), the characteristics of the tropical boundary layer and the mechanisms governing its evolution are revealed by .....including water vapor...."

Line 308. "...inversion has been defeated..." suggest this be changed to "...inversion has been dissipated..."

Ditto Line 310

Line 331 "...in the dry season." Repetitive – not required.

Line 359. "It is since long known..." Sugest change to " It has long been known...."

Line 364. "moves downslope ." This is the first mention of orographic influences on the site measurements. – suggest ">. airflow within the canopy is dominated by orographic effects, in this case leading to mainly local katabatic flows (Aubinet...."

Line 370. "This is useful information when later moving to the diurnal cycle of the vertical particle flux in section..." Suggest this be reworded to "...when discussing..."

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 26881, 2009.