Atmos. Chem. Phys. Discuss., 9, C11455–C11471, 2010
www.atmos-chem-phys-discuss.net/9/C11455/2010/

Atmospheric Chemistry and Physics Discussions

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

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Received and published: 12 March 2010

We thank Anonymous Referee #1 for comments and suggestions for improvement of our manuscript. The comments from the reviewer followed by our responses to the comments can be seen below. All references can be found in the updated manuscript. Changes in the manuscript are marked in red color.

Referee comment 1:

The data is presented in a clear and methodical manner and interpretation and description of diurnal patterns in CO2, 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

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 vd 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.

Response:

Yes, particle size is a key parameter when investigating dry deposition of aerosol particles. Our intention when we started this project was to have SMPS measurements in parallel with the flux measurements for the whole campaign. However, technical problems in particular with the CPCs, caused to a large extent by the very humid and warm conditions in the Amazon rain forest, forced us to prioritize the flux measurements. The SMPS measurements ran without problems in a period between June 13 and July 7 at the same site. This period is just before the period of the dry season flux measurements but it is representative of the dry season size distribution. We have added a section (3.4.1) where the median size distribution (with temporal variability) during the period is presented (Fig. 4) and discussed. Our median size distribution could be described by an accumulation mode, an Aitken mode and a nucleation mode (Fig. 4b), similarly to the wet season size distribution measured by Zhou et al. (2002). The number concentrations, geometrical mean diameters, and geometrical standard deviation of the three modes in both this study, representing the dry season, and in the study by Zhou et al. (2002), representing the wet season, have been provided in Table

3. The log-normal fit together with the median size distribution is shown in Fig. 4b. The Zhou et al. study was based on measurements in Balbina, 125 km north-east of Manaus, in the same part of the Amazon as this study.

Referee comment 2:

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

Response:

The phrasing in the abstract was: "In general, particle deposition velocities in this study are low compared to studies over boreal forests. 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." Thus,

domination of accumulation mode particles and low wind speeds were suggested as explanations for the in general low particle deposition velocities, and not an explanation for the small difference in deposition velocity between the two seasons. (Note that we now use the term "transfer velocity" after comments from referee 2.) However, we agree with the referee that it is important to provide clear information of typical aerosol number size distributions. As was described in the response to the referee comment 1, size distributions representative of the both seasons are now available in Table 3. Concerning mentioning the dominating modes earlier in the paper, the information of the domination of accumulation and Aitken mode particles in the Amazon boundary layer is presented in the introduction. There it is also described that the percentage of accumulation mode particles has been observed to increase during transition from wet season to dry season. However, this is now more carefully discussed in section 3.4.1. We prefer to not provide any information of the dominating particle diameters in the title, since the size distribution cannot be described by only one sentence. We could change "particles" in the title to "sub-micrometer particles", but since we use the term "number flux", we think it should be clear that coarse mode particles will have a negligible influence on a number flux. We have added information in the abstract of the low percentage of nucleation mode particles, so the cited sentence above now reads: "The reasons are probably the high percentage of accumulation mode particles and the low percentage of nucleation 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." In the introduction there was one sentence reading: "Natural biogenic particles are present in both the dry and wet season (Graham et al., 2003), and are a significant fraction of the aerosol mass, with a strong dominance of coarse mode particles (Artaxo and Hansson, 2005)." We have removed the part that discusses particle mass, since this may be confusing in a paper where results from number flux measurements are discussed.

Referee comment 3:

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?

Response:

Yes, the problem with water uptake was a very serious issue during this campaign. It was absolutely necessary to dry the CPC and change butanol at least every second day. When this was not done the measured concentration dropped to zero. This is the primary reason for losses of flux data, as is discussed in the introduction to section 3. Our CPC number concentrations could fortunately be compared with number concentrations from a water-CPC which had a drier installed at the inlet and therefore did not suffer from the same problem. This was a great help when filtering out bad data measured on days when we did not have the possibility to change butanol. The issue of contamination due to high PM loadings was most likely negligible compared to the issue of water uptake and considering the relatively low concentrations.

Referee comment 4:

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, Biogenic secondary organic aerosol will likely by 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?

Response:

It is not likely that new particle formation occurs within the canopy layer. Numbers of nucleation mode particles are low in the Amazon boundary layer. Whereas in other continental locations 3-nm particles are regularly observed at near-surface measurement sites, in the Amazon Basin the smallest particles typically have sizes of 10 to 20 nm (Martin et al., 2008). This has lead to the hypothesis that new particle formation may occur at higher altitudes, which means that the occasionally observed 10-20 nm particles in the surface layer have not likely been formed close to the surface. The observed emission fluxes in the morning during the dry season in this study are neither likely explained by entrainment fluxes, as is discussed in section 3.4.5 in the manuscript. It seems that the most likely explanation is primary aerosol emission from the surface during the night or/and early morning. These particles may stay within the canopy layer throughout the night, since the canopy layer is decoupled from the surface layer above at night, but may be released from the canopy layer in the morning when the canopy is ventilated, similarly to CO2 as is also discussed in section 3.4.5. We of course agree with the reviewer that we cannot provide any information of composition or size of these particles, since we have not performed any measurements of particle composition and size simultaneously with the flux measurements. Therefore we have been careful regarding formulation in the manuscript and used phrases like "it is possible that particles are being emitted" and "these upward fluxes might actually be a result of emission of natural biogenic particles from the forest" instead of using words like "proof". Since these emission fluxes in early morning in the dry season is an issue that definitely deserves more investigation, we still think that the discussion of the emission fluxes should be kept in the manuscript. In general we have tried to make it clearer in the manuscript that the emission fluxes are more likely primary aerosol particles than secondary aerosol particles. The referee is right concerning that if all primary biogenic aerosol particles are supermicron in size, they will have a negligible impact in terms of numbers. However, there are large uncertainties in properties and characteristics of primary biogenic aerosol particles. For instance, the number of fungal spores ex-

isting on Earth is assumed to be in the range of 1-1.5 million (Elbert et al., 2007), but only about 40000 are well-characterized (Rossman, 1994). Furthermore, bacteria are typically 0.25-8 μm in diameter (Thompson 1981) and may therefore make a contribution also in the sub-micron range. Another potential mechanism for generating particle emission is through the plant stomata. Several biogenic related elements (e.g. K, P, S, Zn) in plants are present in the fluids circulating in the plants and can be released from the plant during transpiration (Nemeruyk, 1970). At night time, primary aerosol emission could possibly be related to respiration. Fish (1972) suggested that haze observed in forested areas could be due to submicrometer particles from electrical generation of biogenic aerosol by leaves. Decaying vegetation produce small particles that can act as cloud nuclei (Schnell and Vali, 1973). It has already been shown in earlier studies that the phosphorus concentration increases during nighttime at the lower part of the canopy in the Amazon rain forest. Artaxo and Hansson (1985) and Guyon et al. (2003a and 2003b) attributed this enhancement to nighttime biogenic emissions of particles. The question is whether this emission is only important in terms of particle mass, or if the emission is also significant in terms of particle numbers. We have no proof that the early morning emission number fluxes are explained by primary biogenic aerosol emission, but it seems to be the most likely explanation. However, we have tried to be a little more careful in the discussion of the upward fluxes, and at the end of section 3.4.5 we come up with a suggestion on how it could be shown in the future that the upward fluxes actually represent emission. We have added possible sources of primary biogenic aerosol particles to the introduction and a discussion of why a primary aerosol source is more likely in section 3.4.5.

Referee comment 5:

Furthermore they go to say – line 37/38

“Emitted particles may be stored within the canopy during stable conditions at nighttime, similarly to CO2, 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 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?

Response:

This matter is carefully discussed in section 3.4.5. (section 3.4.4 in the previous manuscript)

“However, the fact that the upward fluxes appear in the morning does not necessarily means that the emission source is lower in daytime than at nighttime. Particles emitted at nighttime may be stored in the canopy layer which is decoupled from the atmosphere in stable conditions. Artaxo and Hanssen (1985) and Guyon et al. (2003a and 2003b), observed an increase in phosphorus concentration during nighttime at the lower part of the canopy, and they attributed this enhancement to nighttime biogenic emissions of particles containing phosphorus. Hence, the upward flux in the early morning is then the flux of approximately all particles that have been emitted and stored under the canopy throughout the night. In daytime, when conditions are more turbulent, an emission of the same magnitude would generate upward fluxes that are more continuous and these emission fluxes would drown in the large daytime deposition flux.”

Referee comment 6:

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 $\pm$ 1σ of the mean.”

Response:

Yes, upward fluxes dominate also in the evening and throughout the night in the dry season. This is particularly clear between 19:00 and 22:00 LT. However, since the early morning peak in upward flux appears simultaneously with the peak in CO2 emission, we consider the morning upward flux peak as the most interesting case. Concerning significance of the flux, we think that between 06:00 and 09:00 LT it can be clearly observed just by looking at Fig. 6 where error bars are represented. However, we have added a case study to the manuscript where some important observations are pointed out. This is presented and discussed in section 3.4.5.

Referee comment 7:

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?

Response:

The test by Foken and Wichura (2007) was not applied to the fluxes in the original manuscript. We now have performed this test by dividing the 30 min periods into subintervals of 5 min. We rejected fluxes when the difference between the flux over 30 min and the mean covariance of the 5 min subintervals exceeded 60 %, as suggested

by Järvi et al. (2009) for particle fluxes. This resulted in a loss of about 36 % of the fluxes. This procedure is now described in the method section. The fluxes have (as in the original manuscript) also been calculated for time scales of 10 and 3 min, as was described in the method section and section 3.4.5, to reduce the risk of mesoscale impact on the flux particularly at nighttime.

Referee comment 8:

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.

Response:

R2 for all equations have been added to the manuscript. We have also added information on how many values the medians and the percentiles have been calculated over in the Figure text. The reason for the relatively large scattering is that both net upward and net downward fluxes contribute to the transfer velocities.

Referee comment 9:

Could the authors provide a plot of particle number flux versus concentration, as discussed 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.

Response:

This would have been a nice method if the background aerosol concentrations were not so high in the dry season. As was discussed in section 3.4.5, the relatively small gain of

particles from the possible emission flux in the morning is insignificant compared to the overall negative trend in concentration throughout the night and in the morning. From section 3.4.5: “an emission source of 0.5ÎĞ106 particles m-2s-1, like the early morning median upward flux in Fig. 6, active during one hour would only increase the particle concentration with 18 particles per cm3 in a $\sim$100 m thick boundary layer, which is only a little more than a one percent increase in particle concentration. The particle concentration in Fig. 8 shows a decreasing trend from midnight until 10:00 LT and the relatively small gain of particles from the emission flux in the morning is insignificant compared to the overall negative trend in concentration. Therefore, particle emission from the forest is still a possible explanation for the median upward flux around 08:00 LT, even though there is no peak in particle concentration at the same time.” This means that the possible emission from the rain forest is very small compared to the relatively high background concentrations in the dry season.

Referee comment 10:

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.

Response:

Yes, it would be preferable to convert our deposition velocities to surface deposition velocities. The reason why this has not been done is that the values of the roughness length and displacement height are associated with large uncertainties for this site. This conversion would produce some new errors. The reason that we still have used some values on the roughness parameters in section 3.4.8, is that we want to show that vd $\approx$ vds. We have added a figure that shows median diurnal cycles of both vd and vds in the same plot (Fig. 12b), only to show that the difference between vd and vds is very small in this study.

Referee comment 11:

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.

Response:

The first question was answered in the response to referee comment 4. The second question, we did not fully understand. If the reviewer means that a downward particles flux prevails in daytime but that these particles stay in the canopy layer and are not deposited, this would still not result in a vertical gradient such that an emission flux would be observed when turbulence increases in the morning. There is no obvious reason that the air above the canopy layer would be diluted at nighttime while the canopy layer is not diluted.

Referee comment 12:

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 vd 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 significant 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.

Response:

Figure 5a and b have now been combined in one figure (Fig. 6). The referee is right concerning the small errors. In the original manuscript we used the counting error to describe the flux uncertainty. However, the counting uncertainty does not include uncertainty due to detrending and non-stationary conditions. In the updated manuscript we now use the random uncertainty suggested by Wyngaard (1973) with the integral time scale from Rannik et al. (2009). The last sentence was discussed in the response to comment 4. We agree that we cannot state that the upward flux is emission of biogenic particles. However, we think we have to discuss that upward particle fluxes are frequently observed simultaneously with CO2 emission fluxes in early morning and try to propose a reasonable explanation.

Referee comment 13:

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 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 vd. 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 vd but typically range from 2.5 – 4.5 mm s-1 (for particle diameters ranging from 0.1 to 0.025 μm) in the former to 6 – 15 mm s-1 (for particle diameters ranging from 0.1 to 0.02 μm) for the latter. In this manuscript (line 525) we are told that the deposition velocities are such that. . . "In both seasons, vd peaks at approximately 1 mm s-1 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-1 in the accumulation mode size range > 0.1 μm. 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 vd have been quoted.

Response:

Yes, this is an important point. We use median transfer velocities in the diurnal cycle, because we want to show what typically happens. We now have made the citation by Pryor et al. (2007) more detailed. In Fig. 4 in Pryor et al. (2007), the median deposition velocity seems to drop below 2 mms-1 already at a geometric diameter of 50 nm.

Referee comment 14:

Figure 1 is rather "fuzzy" and needs to be improved.

Response:

Yes, it looks a bit fuzzy on the ACPD web-site but not in our files. This can probably be solved together with the editor at a later stage.

Referee comment 15:

Line 140. "using a. . . "

Response:

Changed.

Referee comment 16:

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

Response:

The referee is right. However, since we now use the uncertainty as suggested by

Wyngaard (1973) we have instead added this reference.

Referee comment 17:

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

Response:

Changed.

Referee comment 18:

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. . . ."

Response:

We have changed to something very similar to the referee's suggestion

Referee comment 19:

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

Response:

Changed.

Referee comment 20:

Ditto Line 310

Response:

Changed

Referee comment 21:

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

Response:

Changed

Referee comment 22:

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

Response:

Changed

Referee comment 23:

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. . . .."

Response:

Changed

Referee comment 24:

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. . . "

Response:

Changed.

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