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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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We thank Anonymous Referee #2 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.

Introduction comment:

However, parts of the manuscript repeat the previous article by Ahlm et al. (2009) and should be shortened. For example, the "Method" sections are almost identical.

Response:

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Section 2.2 has been shortened. We have also removed the last three sentences of section 2.3.1. We removed the motivation of using the term BCe of section 2.3.3 and instead leave a reference. Equations for correction due to limited time response in section 2.4 have been taken away and we refer to Horst et al. (1997). (However we have added some discussion here according to referee comment 3.) The motivation for not using the Webb correction has been removed and we instead refer to Ahlm et al. (2009). Finally, the "introduction" to section 3 has been shortened.

Specific comments

Referee comment 1:

In my opinion, the abstract is too long and should be shortened. For example, the second part of the first paragraph (The primary goal is to. . .) could be replaced by a summary of the main results and conclusions with regard to these goals.

Response:

We removed the sentences below: "The primary goal is to quantify the dry deposition sink and to investigate whether particle deposition velocities change when going from the clean wet season into the more polluted dry season. Furthermore, it is tested whether the rain forest is always a net sink of particles in terms of number concentrations, or if particle emission from the surface under certain circumstances may dominate over the dry deposition sink."

Referee comment 2:

The authors give emission of natural biogenic particles from the rain forest as a possible explanation for morning upward particle fluxes. Does this imply a primary or secondary source of biogenic particles? What mechanisms could generate these particles? Are there any supporting data available? It is stated from the literature on p. 26884 (lines 21-23) that "natural biogenic particles are present in both the dry and wet season (. . .), and are a significant fraction of the aerosol mass, with a strong dominance of

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coarse mode particles (. . .)". Thus, while coarse natural biogenic particles contribute to aerosol mass, their contribution to aerosol number remains unclear.

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. We agree with the referee that " while coarse natural biogenic particles contribute to aerosol mass, their contribution to aerosol number remains unclear". This was one of the main objectives of this whole project to find out if there is a significant number source of primary aerosol particles in the rain forest. From the results of this study we cannot draw any direct conclusions of composition or source of the particles that are associated with the early morning emission fluxes. However, some potentially important sources in the Amazon rain forest are mentioned here: 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). Fish (1972) suggested that haze observed over forests could be due to submicrometer particles from electrical generation of biogenic aerosol by leaves. Furthermore, decaying vegetation may produce aerosol particles (Schnell and Vali, 1973). Fungal spores are usually in the diameters size range 1-30 μm (Jones and Harrison, 2004), suggesting that they do not contribute significantly to the aerosol number population. However, the number of fungal spores existing on

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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). Finally, bacteria are typically 0.25-8 μm in diameter (Thompson 1981) and may therefore make a contribution also in the sub-micron range. 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 also is significant in terms of particle numbers. We have added these possible sources for 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 3:

The CPC was logged at 1 Hz, and therefore a response time of 1 s has been used to correct the aerosol number fluxes for limited instrumental response according to Eq. 1 in the manuscript. While the time response of the CPC alone may be around 1 s, the measuring system consisting of a 4 m long 1/4-inch sampling line and the CPC with a flow rate of 1.08 l min-1 is certainly much slower. Without a backup flow, the flow regime within the sampling line is laminar and will generate a parabolic flow profile. This will lead to additional attenuation of the number concentration fluctuations. Thus, a slower time response (without knowing the technical details, 1.5 s to 2 s would be my guess) seems to be more appropriate. In addition, the original formulation of Eq. 1 has been put forward by Horst (1997). This reference should be added.

Response:

This is correct. We now have estimated the total time constant of sampling line + CPC. This was done by using transfer equations derived by Lenschow and Raupach (1991) for damping of concentration fluctuations in laminar flow and transfer equations of a sensor (Horst et al., 1997). The total time constant was 1.3 s. The fluxes have been

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re-corrected with this value of the time constant and all CPC flux figures have been re-plotted. We have also added details on this procedure in the method section.

Referee comment 4:

I cannot follow the explanation why the Webb correction has not been applied to particle fluxes. While it is certainly true that temperature fluctuations are dampened in the inlet tubing, I am not sure if the temperature and humidity conditions inside the CPC are relevant considering the time scales when sampling at a frequency of 1 Hz. The vertical advection term that is added to the measured flux when using the Webb correction is evaluated from the water vapor and sensible heat fluxes. One could argue, however, that the Webb correction may be negligible for particle number fluxes because the number fluctuations are typically large compared to the mean number concentrations.

Response:

The Webb correction follows from that ascending air is usually warmer and contains more water vapour, thereby having lower density than descending air. This means that concentration fluctuations measured in the atmosphere also contain a contribution from sensible and latent heat fluxes. However, if the air is for instance cooled down to a constant temperature and also dried before the measurements, the fluctuations of sensible and latent heat fluxes are no longer included in the measured concentration fluctuations. Inside the CPC, the air is cooled down and a significant amount of the water vapour is deposited in the butanol tank before the particles are counted inside the CPC. The temperature and humidity of the air inside the CPC will likely have little "memory" of fluctuations in temperature and humidity in the outside air. In this study we have sampled the condensation temperature inside the CPC but when the particles are being counted and the air flow is set, the temperature will likely have changed. To try to correct for this would only introduce new errors.

As Webb wrote himself in Webb et al. (1980): "It is when the measurement of the constituent's density fluctuations or mean gradients is made in unmodified in situ air that

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corrections arising from the fluxes of both heat and water vapour are required." "If the measurement involves sensing of the fluctuations or mean gradient of the constituent's mixing ratio relative to the dry air component, then no correction is required".

Referee comment 5:

On p. 26890, the authors state that "there are of course other sources of uncertainty in aerosol flux quantification. . .". The authors should state some of the other uncertainties and give at least a qualitative estimate of their relevance for aerosol number fluxes even if it is difficult to quantify their magnitude.

Response:

In the original manuscript we described the flux uncertainty as the counting error. We have now calculated the total random uncertainty according to Wyngaard (1973) and used the integral time scale as suggested by Rannik et al. (2009).

Referee comment 6:

In the discussion of Fig. 2, the authors state that the "curves for sensible (Fig. 2b) and latent (Fig. 2c) heat fluxes are rather well correlated with the PAR". However, both sensible and latent heat fluxes set in 2 hours later. The friction velocity (Fig. 2i) starts to increase 1 hour after PAR starts to increase. These differences should be discussed in more detail because they characterize the morning hours when CO2 and particle emission fluxes were observed. Maybe one could gain more insight if the data were classified on a day-by-day basis in classes when morning particle emission was observed or not. Are there differences in the onset of turbulence (sensible/latent heat fluxes, friction velocity) and the emission fluxes of CO2 between days with and without early morning particle emission? Is the early morning particle emission associated with a certain wind direction?

Response:

Yes, the sensible and latent heat fluxes start to increase about 1.5 hours after sunrise.

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We have added following sentences to the manuscript: “However, the sensible and latent heat fluxes start to increase first ∼1.5 hours after sunrise. This delay might be an effect of negative radiation balance also a while after sunrise.” Concerning the reason that the friction velocity starts increasing (more rapidly) about an hour after sunrise, we have added following sentences: “The friction velocity starts to increase rapidly first one hour after sunrise. The top of the canopy needs to be warmed up before the air temperature above the canopy starts to increase. When the unstable layer reaches the top of K34, about 20 m above the canopy, the friction velocity at the top of K34 is expected to increase. However, since the nocturnal layer is 80-180 m it seems to last until 09:00 LT until the whole nocturnal layer is dissipated (Fig. 2e).” Yes, we have looked at it on a day-by-day basis. What can be said is that the early morning upward particle fluxes often appear simultaneously with the CO2 emission fluxes. Both these upward fluxes appear when the friction velocity starts to increase in the morning. Upward fluxes dominate between 07:00 and 09:00 LT for all represented wind directions. To really prove that the upward flux is a result of ventilation of the canopy, however, one would need a CPC within the canopy layer, to see whether a higher particle concentration is built up there than above the canopy layer at nighttime. Also one would need wind measurements inside the canopy, to see when it is ventilated. Since we do not have this information we cannot prove anything, but since we think this issue deserves more investigation we still think the emission fluxes should be discussed in the manuscript. We have now added a case study (Fig. 9) where some important observations are discussed. On a few days the early morning upward aerosol flux was not observed, even though there was an emission flux of CO2. There was no obvious difference in the meteorological parameters that could explain this. A possible explanation is if the particle concentration would increase in the surface layer during the night while the particle concentration in the canopy layer would remain the same. Then instead downward fluxes would be expected when the canopy is ventilated in the morning. However, this is only speculation and therefore we do not mention this in the manuscript.

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Referee comment 7:

One very interesting aspect of the manuscript is the discussion of the early morning emission fluxes of CO2 and particle number, just when rapid changes in many variables occur. Have the data been tested for stationarity (e.g. Foken and Wichura, 1996)? Did CO2 and particle emission fluxes occur simultaneously, i.e. has particle emission been observed on all days when CO2 emission was observed? When looking at the 75 percentiles, there seems to be a similar feature present in the wet season as well yet less pronounced. Additional information about the atmosphere-canopy coupling regimes would be very helpful. While it is stated that the canopy is decoupled from the atmosphere during stable nighttime conditions, it would be a valuable addition if turbulence data could be evaluated to obtain coupling regimes at the site.

Response:

The test by Foken and Wichura (2007) was not applied to the fluxes in the original manuscript. We have now performed this test by dividing the 30 min periods into sub-intervals 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.

The other questions were answered in the response to referee comment 6.

Referee comment 8:

For calculating the diurnal cycles of the vertical particle flux, data from wind directions between 310 and 20 degrees have been discarded to exclude any impact from the diesel generator and house at K34. Is this based on some kind of footprint analysis?

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Response:

The diesel generator and the house are located in a direction of 340 degrees from K34 at a distance around 2 km. To be sure that the fluxes and concentrations are not influenced by particles transported from the research station, we excluded a quite large wind sector associated with possible transport from the station. This was not based on any foot print analysis, partly because we were not only concerned with removing any emissions fluxes (from within the foot print), but also to avoid particles from this source (even if they originated from beyond the foot print) since they would contribute to concentration and hence to deposition fluxes.

Referee comment 9:

Figure 5a and b present the same data and could be combined in one graph. In addition, I suggest to add error bars to Figure 6 and also to present the uncertainties of the particle deposition velocities in Fig. 8. In addition, some metric to represent the uncertainty in the presented relations of deposition velocity and friction velocity should be included in Eqs. 6 and 7.

Response:

Figure 5a and b have been combined (now Fig. 6). We have added uncertainties to all the curves mentioned by the referee. We have also added R2 values to the relations between deposition velocity and friction velocity.

Referee comment 10:

Particle deposition velocities: Since both particle emission and deposition fluxes can occur and were observed, the term “particle transfer velocity” may be more appropriate instead of “particle deposition velocity”. The authors have used this terminology in their previous manuscript mentioned above. In general, it is difficult to compare studies of particle number fluxes because there is no convention on how to report average transfer velocities. This is discussed by the authors on p. 26900. When averaging upward

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and downward fluxes, the opposite signs of the flux values may lead to unfavorable averaging results. One suggestion is to separate deposition and emission fluxes, i.e. positive and negative particle transfer velocities, and report means or medians of the transfer velocity separately for deposition and emission periods.

Response:

We have changed the term to transfer velocity as the referee suggests, since emission may occasionally contribute to the net flux. In general, there is no perfect way of estimating deposition velocities when both emission and deposition are apparent in the net flux. However, we think it is dangerous to exclude upward fluxes when estimating deposition velocities if it cannot be clearly stated that the upward fluxes are results of a physical process like for instance emission or entrainment. Since the majority of the net upward fluxes most likely are due to random errors, we think that these must be included when calculating the average deposition velocity (or now transfer velocity). We have now described this issue more carefully in section 3.4.6.

Referee comment 11:

The manuscript lacks observational data of the particle size distributions during the dry and wet seasons which is crucial for some of the presented discussions and conclusions. The authors discuss possible differences of the particle size distribution during both periods, but they don't present any direct measurements of the particle size distributions at the site. If such data is not available, it would be necessary to discuss the comparability of the studies cited for particle size distribution measurements in the Amazon and the present study. The influence of dry season biomass burning appears to be less important in the present study. On p. 26895, the authors state that "the Cueiras Reserve is located in an area of pristine rain forest where the direct influence of biomass burning is much lower than in Rondonia or other locations in the southern part of the Amazon rain forest. Even in the dry season, impact of biomass burning emissions is not very high at the Cuieiras Reserve, but can be observed most of the

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time". This contradicts statements such as "during the dry season, when the impact from biomass burning is high" on p. 26904, line 21. Can you give an estimate of the contributions of the Aitken and the accumulation modes to the total particle number concentration during the dry and wet seasons at K34? Some more information about the particle size distribution would be a critical addition to the manuscript.

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 with in particular 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, 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 dry season 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. We have changed from "when the impact from biomass burning is high" to "when the impact from biomass burning is higher" in the Summary and conclusions-section.

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The dependence of particle deposition on stability is compared with a parameterization for sulfate particles by Wesely et al. (1985). Stable cases are discarded because emission fluxes prevail during the nighttime stable conditions, thus not representing the deposition process. This criterion for exclusion seems somewhat random. Emission fluxes may continue throughout the day but be masked by larger daytime deposition fluxes as stated on p. 26900. In this case, the observed apparent particle flux is always a superposition of emission and deposition fluxes, and the parameterization of the deposition flux will be influenced by a potential emission component. Taking this into account and the large variability of the presented fluxes, a straightforward dependence on stability such as the one given by Wesely et al. (1985) may not be expected.

Response:

We agree with the reviewer on this issue. Emission fluxes may occasionally be apparent in both the net upward fluxes and downward fluxes which means that the curves in Fig.10b in the old manuscript may not be expected to agree with the Wesely parameterization. We have removed the comparison with the Wesely-study.

Referee comment 13:

p. 26884, line 1-8: An estimate of the relative contributions of wet and dry deposition to aerosol removal from the atmosphere in the dry and wet seasons would be a valuable addition in this section.

Response:

We could not find any study that quantifies wet removal of aerosol particles in the Amazon basin. Otherwise the suggested comparison between dry and wet deposition in the two seasons sounds like a nice model study for a new paper.

Referee comment 14:

p. 26892, line 8: Replace “what is happening more frequently” by “what is typically hap-

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pening". Since the median is the 50 percentile, "typical" appears to be more adequate than "more frequently".

Response:

Has been changed.

Referee comment 15:

p. 26895, line 4: "Sect. 3.4.2." should be changed to "Sect. 3.4.3."

Response:

Has been changed to sect. 3.4.4 since we have added a section of size distributions (3.4.1).

Referee comment 16:

p.26895, lines 12-13: ". . .dry season particle concentration most of the time was three times higher. . .": Suggest to change to ". . .dry season particle concentration was typically three times higher. . .":

Response:

Has been changed.

Referee comment 17:

p. 26901, lines 1-3: For comparison, some typical values of deposition velocities over boreal forests (and maybe also other environments, e.g. marine environments) may be a valuable addition.

Response:

We have extended the discussion of previous studies and compared with size-resolved dry deposition velocities from Pryor et al. (2007).

Referee comment 18:

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p. 26901, line 18: Eq. 7 slightly deviates from the relation of deposition velocity and friction velocity during the wet season previously published as Eq. 9 in Ahlm et al. (2009). What is the difference in data reduction and which of the two relations is recommended for further use?

Response:

The reason for this is that Ahlm et al. (2009) focused on the cleanest conditions in the wet season and excluded data with higher number concentrations and black carbon concentrations than certain limits, in order to study pristine conditions. Which to recommend depends on the application.

Referee comment 19:

p. 26903, line 19: ". . . it is obvious from Eq. (9)" should read ". . . it is obvious from Eq. (10)".

Response:

This section has been changed.

Referee comment 20:

p. 26904, line 1: remove second "on L-1"

Response:

No longer in the manuscript.

Referee comment 21:

p. 26914, Tab. 2: The median of the diurnal min of BCe concentration is missing and should be added.

Response:

Has been added.

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