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Interactive comment on “Emission and deposition of accumulation and coarse mode particles in the Amazon basin” by L. Ahlm et al.

Anonymous Referee #1

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General comments:

Ahlm et al. present and discuss eddy covariance measurements of aerosol number fluxes over the Amazon rain forest obtained with an optical particle counter. This manuscript is the third in a series of articles by the authors on aerosol number fluxes in the Amazon basin from the Brazilian-Swedish AMAFLUX project. It focuses on size-resolved particle flux measurements in the accumulation and coarse mode, and the authors discuss the diurnal cycle of aerosol concentrations and fluxes, and their dependence on wind direction, anthropogenic influence and potential primary biogenic emission. Since there is very little observational data about size-resolved aerosol fluxes, the topic of the paper is highly interesting. However, the paper does not give sufficient information about the eddy covariance setup, and the data reduction procedures are

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incomplete. I have several major concerns that should be taken into account by the authors:

1. When size-resolved aerosol number fluxes are measured, deliquescence and hygroscopic growth must be taken into account if the particles are not measured under dry conditions (e.g. Kowalski, 2001). A given aerosol number size distribution will vary with changing relative humidity, and this will affect the eddy covariance fluxes, most often inducing a positive covariance. This may be perceived as an upward flux which may be interpreted as particle emission. The authors did not measure the particle size distribution at ambient humidity conditions, yet from my understanding they did not completely eliminate humidity fluctuations. In section 2.3.1, the authors describe their drying system as 1:1 diffusion of particle free air with zero humidity. While this procedure reduces the ambient humidity fluctuations in the sample flow, it does not fully eliminate humidity fluctuations. Even if the influence of hygroscopic growth is small (e.g. Kowalski (2001) discuss influences at relative humidities as low as 15 %), the effect on the calculated fluxes can be large if the slope of the size distribution is steep in the range covered by the OPC (e.g. Vong et al., 2010). This is the case in this study (cf. Fig. 3a and b), and it is absolutely essential to evaluate the influence of hygroscopic growth on the flux calculations. Please take this correction into account and give an estimate of the particle size change in your drying system.
2. Given the low number of particles and the associated uncertainty due to counting statistics, it is absolutely essential to provide some more information on the optical particle counter (OPC) measurements. How did you arrive at a response time of 1 s of the OPC? Is this the 1/e response time or the 95 % response time of the counter? Did you experimentally validate the 1.2 s time constant of OPC + sampling line? Please show some typical power spectra of the particle number time series (both for total particle number in the OPC size range, and for some of the size-resolved particle number time series). The power spectra will help to evaluate the influence of noise on the flux estimates. In particular, the spectral slope in the inertial subrange will give the

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reader a better idea about the flux dampening due to the laminar flow in the sampling lines and the limited sensor response. In my opinion, the spectral analysis of size-resolved particle number time series is a crucial addition to the paper.

3. All rainfall periods have been excluded from the presented results. Does this bias the diurnal patterns of particle concentrations and fluxes especially in the wet season period? Can you show the diurnal patterns of relative humidity and atmospheric stability in order to give a better picture of the potential influence of humidity and suppressed turbulence on the vertical particle exchange. More information about relative humidity would also contribute to a more complete discussion of the potential influence of humidity on primary biogenic particle emissions such as active discharge of fungal spores.

4. In section 3.5, the authors present an equation relating the flux of the 0.5–2.5 μm particles and the horizontal wind speed, and propose to use it to describe the emission flux in models. However, the measured net emission flux is a combination of emission and deposition of particles, and can only be an approximation of the primary particle emission flux. While wind speed may show a slightly better correlation with the net fluxes than friction velocity, I don't think one can draw the conclusion that wind speed could be a key parameter for emission fluxes of 0.5–2.5 μm particles given the large uncertainties of the particle flux estimates. Taking the discussion on triggering mechanisms of particle emission into account, one cannot even expect a moncausal relationship between emission fluxes and wind speed. Thus, this section should be carefully revised.

Additional comments:

5. I think that Figure 1 is not necessary and that it can be removed from the manuscript.
6. Eq. 1 is not consistent with Crane and Evans (1977), where the impaction efficiency E equals the Stokes number times half the bend angle. Also, please correct the citation in the list of references.

7. In section 2.4.2, can you add the 25 and 75 percentiles (or 10 and 90 percentiles) of the flux uncertainty due to counting statistics. The interquartile range is a good representation of the spread of uncertainty due to counting statistics.

8. A maximum in the second size channel can be observed in particle number and volume in Fig. 3 a,b,c. Can you speculate if this is a real maximum or if the particle counts in the lowest size channel may be too low?

9. I cannot entirely follow the discussion of Fig. 4a. The authors observe a vague trend "of slightly decreasing concentrations during the morning in the dry season, but increasing concentrations during the morning in the wet season." Is this observation restricted to the period from 08:00 to 10:00 local time? This is the only period where I can see an increase in concentration in the wet season curve. On the other hand, the decreasing trend in the dry season can be observed more or less from midnight until noon. Taking this into account, I cannot follow the conclusion that mixed layer growth and associated entrainment on average may have a diluting impact on particle concentration in the dry season.

10. In the discussion of Fig. 5, the authors state that the two largest channels are highest between 00:00 and 03:00 local time in the wet season. To me, it looks like the time period from 23:00 to 02:00 shows the highest concentrations. Furthermore, the normalized presentation of the particle concentrations makes it difficult to take into account the uncertainty of the measurement especially in the larger size channels.

11. In Fig. 10b, emission fluxes of coarse mode particles can be found in the wet season from sunrise through the evening, with a maximum in the afternoon at 15:00 local time. In Ahlm et al. (2010), upward fluxes (probably dominated by particles < 100 nm diameter) in the early morning hours of the dry season were presented and interpreted as primary biogenic particles, emitted and stored under the canopy at nighttime. Does this imply different emission mechanisms of primary biogenic particles in different size ranges, or does this imply different turbulent transport processes in the

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wet and dry seasons, or is it a combination of emission, transport, and maybe other processes?

12. In section 3.5, transpiration from plants is mentioned as a potential mechanism for particle emissions, but discarded due to the fact that the latent heat flux is not related to the "emission" flux. It should be made clear that the latent heat flux is not a direct measure of transpiration but also influenced by evaporation and other processes.

Some minor corrections:

- p. 14015, l. 15: Replace "isoprenes" by "isoprene".
- p. 14016, l. 4: Replace "makes" by "make".
- p. 14031, l. 17: Replace "equaton" by "equation".
- p. 14034, l. 24: Insert a space between "U" and "is".

References:

Ahlm, L., Nilsson, E. D., Krejci, R., Mrtensson, E. M., Vogt, M., and Artaxo, P. (2010) A comparison of dry and wet season aerosol number fluxes over the Amazon rain forest, *Atmos. Chem. Phys.*, 10, 3063-3079, doi:10.5194/acp-10-3063-2010.

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