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.

Referee comment 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.

Response:

The aerosol particles in the Amazon rain forest are strongly dominated by organic compounds. Gunthe et al. (2009) estimated the organic mass fraction to 90 % at the same site as this study. Rissler et al. (2006) has investigated hygroscopic properties of the aerosol in the Amazon and found no step-like deliquescent behavior of the aerosol in the dry or wet period. Nevertheless, it is true that one has to consider hygroscopic growth when measuring size-resolved particle number fluxes. However, in this study hygroscopicity has a negligible impact on the results. The most important reason for this is that we have dried the aerosol before sampling.

Our drying procedure, described in the manuscript, removes 50 % of the water vapor (of the air entering the inlet, 50 % is completely dried and later mixed with the other 50 %, that is not dried, before sampling in the OPC). This means that the relative humidity before sampling decreases to 50 % of its atmospheric value (since the partial pressure of water vapor is directly proportional to the number of moles of water vapor).

Rissler et al. (2006) found that the hygroscopic growth factor of aerosols over the Amazon rain forest could be described by following equation:

$$Gf = \left[1 + A \cdot \left(\frac{RH/100}{1 - RH/100}\right)\right]^{1/3} \qquad (1)$$

where RH is the relative humidity. The factor A had the value ~0.1 for the most hygroscopic particles during the wet period (D_P > 250 nm). Eq. 1 gives a growth factor of ~1.24 in 90 % relative humidity. However, in daytime, where we observed our largest emission fluxes in this study, the atmospheric

relative humidity was on average 70-80 % (Fig. 16b in updated manuscript). After the drying procedure, the corresponding relative humidity will be 35-40 %, which gives a growth factor of only 1-2 %. Hence, hygroscopic growth seems to be negligible in this study. Even though the relative humidity was occasionally close to 100% in daytime on a few days (or more frequently at nighttime), the corresponding 50 % relative humidity after drying only gives a growth factor of ~3%. We have added some information on this issue in the method section.

Referee comment 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 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.

Response

The time constant that is necessary for estimating the flux attenuation at higher frequencies, according to Horst et al. (1997), was obtained from the relation $c_m(t) = c_2 + (c_1 - c_2)e^{-t/\tau}$, where τ is the time constant (Doebelin, 1990). This relation describes a step change in concentration from c_1 to c_2.
The time constant was provided to us through communication with Grimm.
However, we have now estimated τ at Stockholm University, by using data from Grimm, and we get a much lower value on τ. In the setup, a zero-filter was used and the OPC response time was estimated for a step decrease to zero concentration, rather similar to the setup by Quant et al. (1992) and Buzorius (2001). We have estimated τ to ~0.3. The experiment was repeated several times with very similar results. Therefore, we believe that the estimation is reliable.
We think that there was some misunderstanding between us and the person we talked to at Grimm, and what we received from him might have been the time it took for the concentration to drop to zero, and not the actual time constant.
However, we now use τ=0.3 for the OPC.
To estimate the total time constant, OPC + sampling line, we use cospectral transfer functions for a sensor (Horst et al., 1997) and for a tube (Lenschow and Raupach, 1991) and multiply these. The resulting transfer function is compared with the Horst et al. transfer function with a value on τ that fits the total transfer function. We have described this more carefully in the updated manuscript and added Fig. 1, which illustrates this procedure. The magnitudes of the corrections have been updated in the method section. The flux figures and Eq. 9 in the manuscript have also been updated.
We have also added some examples on variance and covariance spectra (section 2.5) for the total OPC size range and for particles in the diameter range 0.5-2.5 μm. It is difficult, though, to draw conclusions of flux attenuation from looking at spectra in this study. The level of noise was highly

variable at different variance spectra. However, even when the noise level was relatively high in the variance spectra, the co-spectra looked generally well, indicating that the noise was not correlated with vertical wind and thereby not significantly affecting the flux.

Referee comment 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.

Response:

There is no significant bias when excluding half hours with rain. The reason for this is likely that all presented diurnal cycles represent median values. Even though rain amounts are large in the wet season, there are very few half hours with rain compared to half hours that are rain free, even in the afternoon.
We have added median diurnal cycles of stability (Fig. 5d) relative humidity (Fig. 16b), as well as some other meteorological parameters (Fig. 16).

Referee comment 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 monocausal relationship between emission fluxes and wind speed. Thus, this section should be carefully revised.

Response:

It is true that the measured flux is the sum of emission and deposition. It would be desirable to be able to correct for dry deposition in order to obtain the "real" emission. This is sometimes done by using deposition models like Slinn (1981) or Zhang et al. (2001). However, from our experience of using these models in the past, we know that they are very sensitive to parameters like roughness length, displacement height, sizes of collectors, wind speeds at reference heights and drag coefficients. All these parameters are highly uncertain for the site in this study. Therefore, we are afraid that an attempt to correct for dry deposition would likely increase the uncertainty in the emission rather than decrease the uncertainty. However, we have made it clearer in the manuscript that the emission described by Eq. 9 also include deposition and is therefore likely underestimated.

Regarding correlation to wind speed/friction velocity, we agree that the uncertainties are large in these types of measurements. However, we think it would be a mistake to not come up with the best possible parameterization based on the data we have. We have been very clear, in the manuscript, with that the fact that the upward fluxes are well correlated with horizontal wind speed does not necessarily mean that wind speed is what triggers the emission. We have used formulations in section 3.5 as:

"From this study it cannot be stated whether the wind is only a transport mechanism out of the canopy layer, or if the wind (and the turbulence it creates) has a direct impact on the actual emission from the specific source."

and

"Thus, the observed emission fluxes of particles in the diameter range 0.5-2.5 μm indicate a source of biogenic particles in the rain forest, but more specific source mechanisms cannot be defined from this study."

We changed following sentence in the original manuscript

"Actually, both peaks in emission particle flux (at 11:00 and 15:00 LT) appear at exactly the same time as the peaks in wind speed, indicating that wind speed could be a key parameter for emission fluxes of 0.5-2.5 μm particles"

to following sentence were we avoid the words "key parameter"

"Actually, both peaks in emission particle flux (at 11:00 and 15:00 LT) appear at exactly the same time as the peaks in wind speed, indicating that the net emission flux is correlated with horizontal wind speed."

Referee comment 5:

I think that Figure 1 is not necessary and that it can be removed from the manuscript.

Response:

We have removed that figure, and also slightly rewritten the following sentence in section 3.3, that in the original manuscript was referring to Fig. 1:
"In fact, the minimum concentration is found in the north-westerly wind sector, the specific direction to the diesel generator at the research station."

Referee comment 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.

Response:

Okay, we took the equation from Hinds et al. (1999) since we did not have the original paper by Crane and Evans (1977). We trust the referee in that it should be half the bend angle, and we have updated the equation values of the inertial losses in the method section of the manuscript.

Referee comment 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.

Response:

We have added 25 and 75 percentiles in Fig. 2 and in section 2.4.2 in the updated manuscript.

Referee comment 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?

Response:

We have no reason to believe that the first channel would show incorrect concentrations.
Pöschl et al. (2010) presents a size distribution measured by DMPS during the wet season with the accumulation mode centered around 200 nm. The dry season size distribution measured by DMPS had an accumulation mode centered around 150 nm. So, it seems as the peak should be located slightly at a slightly lower diameter than 250 nm where the OPC starts measuring. However, OPC and DMPS measurements are not always perfectly correlated. We cannot give a definite answer to this question from the measurements in this study.

Referee comment 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.

Response:

We agree with the referee that the trends are small in that figure. Therefore we have removed the sentences from line 26 on page 14023 to line 7 on page 14024 (in the old manuscript). To make it consistent, we have also removed similar conclusions on lines 8-15 and line 17 on page 14025.

Referee comment 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.

Response:

We have changed to 23:00 to 02:00. The reason for normalizing in this way is to be able to show the diurnal variations for all channels. If we would show absolute concentrations, bins representing the larger particles would not be seen in the plot. This was the case even when we tried with logarithmic concentrations. The figures give good information on the diurnal cycles of different sizes of particles, in a percentage point of view. To get the absolute concentrations, one has to look at table 1.
Counting errors are very low for these concentration measurements, from ~0.1 % for the smallest particles to ~2 % for the larges particles.
The figure of course gives no information of day to day temporal variability, but we cannot present plots for all size bins separately. This figure was meant to give some information on the fact that particles with different sizes have different diurnal trends, before focusing on fluxes of different particle sizes in the later sections.

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

Response:

We have added the paragraph shown below to section 3.4:

“That the emission peaks during afternoon also suggests that the source here cannot likely be related to the potential emission observed as upward particle fluxes during early morning in Ahlm et al. (2010). However, that study focused on the total aerosol number population (D_p > 10nm) whereas the emission fluxes discussed here only include particles in the diameter range 0.5-2.5 μm. The contribution from this interval makes a negligible contribution to the total number concentration studied in Ahlm et al. (2010). Therefore, there is no contradiction between the observed emission

fluxes here, peaking in the afternoon, and the early morning upward fluxes observed in Ahlm et al. (2010).”

In the Ahlm et al. (2010) study, the hypothesis was that increasing wind or turbulence in the morning ventilated the canopy resulting in upward fluxes of particles that had been emitted and stored in the canopy throughout the night, similarly to what has been observed for CO_2. In this study, however, we cannot tell whether the wind is only a transport mechanism out of the canopy layer, or if the wind actually triggers the emission inertially. We have added following paragraph (the end of section 3.5) as an outlook for what measurements that should be done in order to learn more about the source and source mechanism:

“To further investigate the actual emission process, particle number fluxes should be measured simultaneously in the canopy and above the canopy, similar to the study by Grönholm et al. (2009). These types of measurements could reveal whether the wind actually triggers the emission, or if the correlation between upward particle fluxes and wind speed results from storage and ventilation processes. Additional aerosol chemistry measurements inside the canopy would increase the possibility to determine what types of particles that are emitted and what sources are responsible for the emission.”

Referee comment 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.

Response:

We have included that information.

Referee comment 13:

p. 14015, l. 15: Replace "isoprenes" by "isoprene".

Response:

We have changed this.

Referee comment 14:

p. 14016, l. 4: Replace "makes" by "make".

Response:

Changed.

Referee comment 15:

p. 14031, l. 17: Replace "equaton" by "equation".

Response:

Changed.

Referee comment 16:

p. 14034, l. 24: Insert a space between "U" and "is".

Response:

Changed.

References not mentioned in the manuscript:

Buzorius, G.: Cut-Off Sizes and Time Constants of the CPC TSI 3010 Operating at 1-3 lpm Flow Rates, Aerosol Science and Technology, 35, 577-585, 2001.

Gunthe, S. S., King, S. M., Rose, D., Chen, Q., Roldin, P., Farmer, D. K., Jimenez, J. L., Artaxo, P., Andreae, M. O., Martin, S. T., and Pöschl, U.: Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity, Atmos. Chem. Phys., 9, 7551–7575, 2009.

Pöschl, U., Martin, S. T., Sinha, B., Chen, Q., Gunthe, S. S., Huffman, J. A., Borrmann, S., Farmer, D. K., Garland, R. M., Helas, G., Jimenez, J. L., King, S. M., Manzi, A., Mikhailov, E., Pauliquevis, T., Petters, M. D., Prenni, A. J., Roldin, P., Rose, D., Schneider, J., Su, H., Zorn, S. R., Artaxo, P., and Andreae, M. O.: Rainforest Aerosols as Biogenic Nuclei of Clouds and Precipitation in the Amazon, Science, 329, 2010.

Quant, F. R., Caldow, R., Sem, G. J. and Addison, T. J.: Performance of Condensation Particle Counters with Three Continuous-Flow Designs, J. Aerosol Sci., 23, 8405-8408, 1992.