

We thank Anonymous Referee #3 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:

One of my main concerns about the paper is that the information of what is basically an integrated study is spread rather too thinly across too many papers, which makes it more difficult to provide a coherent, comprehensive picture of the emission / deposition processes at the site. I would therefore strongly encourage the authors to include the further material on the deposition into this paper, rather than splitting it off into a separate publication, for reasons that will become apparent below.

Response:

We agree with the referee that including more information on deposition gives a more complete picture. The reason why this was not done in the original manuscript is that we thought such a paper would be too long, why we felt that we had to split the results into two papers. However, we have now followed the referee's suggestion and included the main results and conclusions of what was meant to be a separate paper. These results can now be found in section 3.3.2. Adding these results also meant that it was necessary to update the abstract, the introduction and the summary/conclusions sections.

Referee comment 2:

In disagreement with comment 1 of Referee #1 I believe that the drying of the sample stream has been the key to these measurements succeeding where others have failed. If the dryer is sufficiently effective for the OPC to measure the dry radius, the corrections implied by Referee #1 clearly do not apply. However, it would be worth showing (possibly through a simple modelling exercise or through a test with two dryers in series), how much of the water is likely to have been removed in the setup.

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:

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

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 3:

Related to this point is the fact that equilibration with water vapour is not the only potential source of artefacts on the measurements. Loss or uptake of semi-volatile aerosol pre-cursor gases (organic and inorganic) can equally result in particle growth or shrinkage that leads to spurious number fluxes for fixed-sized bins as e.g. pointed out by Nemitz and Sutton (2004) and Nemitz et al. (2009) for the NH_4NO_3 equilibrium. This should at least be noted in the revised manuscript.

Response:

This is now noted in the method section and we have added these two references by Nemitz and Sutton (2004) and Nemitz et al. (2009).

Referee comment 4:

Expanding on comment 4 of Referee #1, it is true that the setup measures the (bi-directional) net flux, which is potentially composed of emission and deposition. In order to derive robust source functions for the aerosol (Eq. 6) the fluxes should be corrected for deposition, unless this can be shown to be negligibly small. Similarly, for the quantification of deposition velocities, the ‘contamination’ by emission processes may need to be considered. In my mind, if emission and deposition were fully considered in the same paper a more thorough job could be done in correcting Eq. (6) for the deposition term. Incidentally, what is the effect of gravitational settling on Eq. (6)? It really depends on whether the authors want to derive the source function or the net flux from the canopy, although the former would be more informative. With reference to the literature on resuspension I support the use of U (rather than u^*) to parameterise the emission flux. However, I am surprised that the relationship between U and u^* (Fig. 11) appears to be quite variable at this site. What is the reason?

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 increase the uncertainty in the emission rather than decrease the uncertainty.

Another way of correcting for dry deposition would be to investigate the deposition of particles in the diameter range 0.5-2.5 μm in the anthropogenic sector (the sector that is excluded when analysing emission) as a function of horizontal wind speed. Then these deposition fluxes could be added to the emission function described by Eq. 9. We have looked at the fluxes in this sector and how they depend on wind speed. In fact, the fluxes in the anthropogenic sector are dominated by deposition, though smaller than the emission fluxes in the clean sector. The problem is that these net deposition fluxes are likely to contain the same emission as is observed in the clean sector. Therefore, it would be no true correction to add these net deposition fluxes to the net emission fluxes in the clean sector. To what degree, emission is cancelling deposition in the anthropogenic sector is impossible to know.

Therefore, we think the best way is to present the emission as Eq. 9. However, we have made it clearer in the manuscript that the emission described by Eq. 9 also include deposition and is therefore likely to be somewhat underestimated.

Concerning gravitational settling, the settling velocity v_{TS} can be approximated by

$$v_{TS} = 3 \cdot 10^{-5} d^2 \text{ m/s for } 1 < d < 100 \mu\text{m} \quad (\text{Hinds, 1999})$$

where d is given in μm . This gives a settling velocity of $2 \cdot 10^{-3} \text{ mms}^{-1}$ for particles with a diameter of 0.25 μm , and 0.2 mms^{-1} for particles with a diameter of 2.5 μm , which represents the whole OPC size range. These velocities are much lower than the observed daytime transfer velocities in the 0.25-0.45 μm particles in this study. For the larger particles in the OPC size range, the deposition velocity can be expected to be even larger. Therefore it seems that gravitational settling is negligible for the particles in the OPC size range, at least in daytime.

It is true that U and u^* are not perfectly correlated. One reason could be that most of the turbulence in the convective boundary layer in the tropics is generated by buoyancy rather than wind shear. Horizontal wind speed is often low in daytime, but turbulence is generated anyway because of the unstable stratification.

Referee comment 5:

Like Referee #1 (their comment 11) I was left wondering how the results of the coarser particle fluxes tie in with the observed morning emission peak in total particle number fluxes (Ahlm et al., 2010), which are used to motivate this analysis (page 14015, lines 27ff). It appears that the temporal pattern in the total number flux is not consistent with the biogenic primary aerosol emission implied by the measurements here and thus, a different explanation needs to be found for this phenomenon, which has now also been reported for another tropical forest (Whitehead et al., 2010). The implications of the findings for the earlier publication need to be discussed.

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 ($D_p > 10\text{nm}$) 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).”

Referee comment 6:

Title: In the air pollution community the term ‘coarse mode’ usually refers to the size range 2.5 to 10 μm , whereas this paper deals with particles in the size range 0.25 to 2.5 μm . I would therefore suggest amending the title to bring into line with common terminology.

Response:

The term “coarse mode” is sometimes referred to as particles larger than 1 μm . However, we have followed the suggestion of the referee, and thereby changed the title to “Emission and dry deposition of accumulation mode particles in the Amazon basin”.

References not mentioned in the manuscript:

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.