

Interactive comment on “Aerosol number fluxes over the Amazon rain forest during the wet season” 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.

Referee comment 1:

I am troubled, however, by two ways in which the discussion is presented when the terms "secondary aerosol particles"(or "natural" or "primary"; lines 29, 30, 32, 86, 494, 624, 649, 669, 693) and "advection" (lines 534, 639, 648) are utilized. It seems to this reviewer that there is no evidence that the particles are primary or secondary here (or natural vs. anthropogenic) in that their formation mechanism is not able to be

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inferred other than by simply ASSUMING that the usual processes occur upwind. What the study does suggest is that local production of new aerosol (within the upwind flux footprint) is limited due to a lack of frequent upward fluxes but it does not show how the particles that often mixed downward with entrainment were originally formed. (There is no way to distinguish particle production reactions that occur upwind versus direct emission of particles upwind). All references to secondary or primary or natural ought to be removed from the text. This will not substantially limit the authors' conclusions that fluxes usually are downward to forests for this situation.

Response:

Since the primary goal of this study was to investigate whether the Amazon rain forest provides cloud condensation nuclei (CCN) to the atmosphere by primary aerosol emission, similarly to sea salt emission from oceans, we are of course very concerned of keeping the discussion of primary/secondary aerosols. However, we think the reviewer has some important points that deserve attention. First of all, if the Amazon rain forest-atmosphere system was a closed system with a homogenous surface and all particles were primary aerosol particles, net particle fluxes would on average point upward in the surface layer. There would not be a balance between primary aerosol emission and dry deposition. The reason for this is that wet deposition is a large fraction of the total deposition over the rain forest during the wet season. However, the rain forest-atmosphere system is not a completely closed system even in the wet season, which will be discussed further down. What can be stated from this study is that we did not observe any significant number source of primary aerosol particles within the footprint of the measurement tower during this wet season campaign. Downward fluxes dominated and upward fluxes were to a large extent associated with entrainment. The suggestion by the reviewer that no conclusions may be drawn whether the deposited particles are secondary particles or primary aerosol particles emitted from outside the footprint is an interesting comment that leads to the question how horizontally homogenous the rain forest is concerning primary aerosol emission from the forest. Part of our arguments

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is based on a relatively homogeneous rain forest regarding primary aerosol sources. It is of course a possibility that the Amazon rain forest is not horizontally homogeneous concerning primary aerosol emission. This no one can answer today. However, there are indications that Amazon rain forest is a rather homogeneous entity with respect to aerosol sources and sinks. Previous observations of aerosol number concentrations and size distributions [e.g. Guyon et al., 2003; Krejci et al., 2003; Rissler et al., 2004; Roberts et al., 2001; Zhou et al., 2002] show very stable aerosol number density and aerosol size distribution with very low variability in space and time during the wet season. This is a rather strong argument for horizontal homogeneity concerning rain forest properties with respect to sources and sinks of aerosol particles. Of course, we are still far away from a complete picture of aerosol dynamics, sources and sinks over the rain forest, but so far there is no indication of large variability during natural conditions undisturbed by biomass burning. The motivation for making flux measurements is often that the footprint of the tower can represent a larger area than for example measurements based on exchange over a single branch of a tree. For instance, the objectives of the ongoing CO₂ flux measurements over the Amazon rain forest are to quantify the surface-atmosphere exchange of CO₂ over the whole rain forest, not only within the footprint of the measurements. The same goes for aerosol flux measurements in marine conditions with the goal of quantifying sea salt emission over ocean, not only the footprint of the measurements. This study is to our knowledge the first peer-reviewed published study ever of direct measurements of aerosol number fluxes over the Amazon rain forest, and therefore the only piece we have so far. To draw no conclusions of the sign of the flux only because this is the first study, we think would be a mistake. Until aerosol exchange gets the same attention as CO₂ exchange has in certain networks, we will have to work with single point flux measurements. We have been very clear with that more flux measurements need to be made in other areas of the Amazon rain forest before any final conclusions may be drawn concerning the importance of primary aerosol emission as an aerosol number source in the Amazon rain forest. Until then, it is reasonable to investigate in the discussions what conclusions that would be valid

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if our flux measurements are representative of at least a large part of the surrounding rain forest beyond the footprint. Still we think that the discussion the reviewer opens up of possible horizontal variations of primary aerosol emission from the rain forest is important, and deserves attention. Therefore we have added this as an additional explanation to the hypothesis that the downward flux may be explained by secondary aerosol particles. This means that we through the whole manuscript now are more careful concerning linking the downward particle flux in the cleanest conditions to secondary aerosol particles. What we now state in the abstract is that the number source of primary aerosol particles within the footprint of our measurements was small in this study. We have withdrawn the earlier statement that the “impact of primary aerosol emission on the aerosol number population was small”. “would” has been changed to “could” at line 86 and we have added entrainment as a process that may produce upward particle fluxes in the introduction. In section 3.8, we have made similar changes and included a discussion of possible horizontal variations in primary aerosol emission. The reviewer also suggests in comment 1 that no conclusion can be drawn whether the particle flux in this study is controlled by natural or anthropogenic particles. However, this is exactly what is investigated in Fig. 14. Fig. 14b illustrates how the particle transfer velocity changes when going from higher concentrations of equivalent black carbon (BC_e) to extremely clean conditions. The particle transfer velocity is positive (downward flux) even in the absolute cleanest conditions. Hence, downward particle fluxes prevail even in the cleanest conditions obtained in this study and therefore cannot be explained by deposition of anthropogenic particles. We can understand that the reader may have an impression when reading this manuscript that the location of the measurements is a rather polluted area, because so much effort is spent on describing the data filtering process. However, it must be stated that the atmospheric boundary layer over the Amazon rain forest in the wet season represents among the cleanest continental conditions that can be found on Earth, close to aerosol number concentrations over remote oceans (Andreae et al., 2008). These low concentrations, however, make it important for us to remove any trace of additional anthropogenic aerosol num-

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ber when analyzing the relatively small resulting aerosol number fluxes. One of the key motivations for wet season field measurement campaigns in the Amazon basin during the last 20 years has been to use the basin as a laboratory to study pristine continental aerosol particles (Martin et al., 2009). Background aerosol properties have been studied before at the specific location of this study (Gunthe et al., 2009; Prenni et al., 2009) and at the Ducke Forest Reserve, located about 40 km SE of the Cuieiras Reserve, (Artaxo and Maenhaut, 1990; Artaxo et al. 1990). The reason that we still make so much effort in this study filtering out possible pollution events and inflow of mineral dust is that we want to make sure that the downward particle flux prevails when the flux is controlled by natural particles within the rain forest-atmosphere system. We have added some information (in the beginning of section 3.3) about the close to pristine conditions that prevail over the Amazon rain forest in the wet season. We think this is useful background information before describing the filtering of data that may have been influenced by pollution.

Referee comment 2:

The last paragraph of the abstract is not useful and should be deleted.

Response:

We have deleted the last sentence of the abstract.

Referee comment 3:

The other misused term here is "advection", which the authors use very generally to imply transport into the region by the mean wind but in an eddy flux sense this is a specific term in the budget equation which, if non-zero, of necessity would imply that time change (i.e. non-stationary; storage; as analyzed in Fig.10 here) and/or change in flux with height MUST occur (see J.A. Businger in J. Climate Applied Meteorol. 25, 1101, 1986 and other work). IF there is a change in flux with height the measurements presented here have much less value because they are not representative of any larger

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area. Thus, "advection" is a term to use very carefully with eddy fluxes (see also G.Slinn, "Precip. Scavenging, Dry Depos....", 1983). Thus the statement in lines 144-146 is NOT correct when advection and/or time change (in the sense of the eddy flux budget equation) are important. The authors present no citation for this statement which is o not correct despite the many times that it is ASSUMED in order to simplify a study.

Response:

We agree with the reviewer that the storage and advection terms must be small for the measured vertical flux to correspond to the surface flux. We have removed the statement in lines 144-146, since this statement is not exactly true. However, at least in daytime we expect to have relatively stationary conditions in the data that remains after the data filtering process described in section 3.3. However, earlier studies have shown that the advection terms may play a larger role at nighttime in relative to the measured turbulent flux (Marcolla et al., 2005; Araújo et al., 2008). Therefore, we of course expect the nighttime fluxes in our study to be less reliable than the daytime fluxes. This is the reason why we added a friction velocity-filtered diurnal cycle in Fig. 9d to the manuscript. (However, we found an error concerning Fig. 9d. This figure has been updated.) The fact that nighttime fluxes are less reliable was mentioned in the manuscript already before, but we now have made this point more clear both in the CO₂ and particle flux sections. We have also added a discussion of drainage flows at nighttime. However, nocturnal drainage flows and advection are much worse problem in the dry season when nights are often clear and the stratification is highly stable. In the wet season, the nocturnal stratification is less stable and turbulence is not suppressed to the same degree as in the dry season. The most critical parameter with most variability is certainly the aerosol number concentration. Still the aerosol number concentration and size distribution are relatively stable in the Amazon boundary layer during the wet season. The size distribution has two well defined modes, an Aitken mode and an accumulation mode. Numbers of ultrafine particles are very low (Rissler

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et al., 2004; Zhou et al., 2002). In this study, more rapid increases in aerosol number concentration could occasionally occur when the wind direction was SSE with transport of particles from Manaus. However, this was not the dominating wind direction in this study and these pollution events have been excluded from the data set in the data filtering process. Other rapid changes in aerosol number concentration may occur during rainfall. However, also these events have been excluded from the data set. This point, that the filtering process strongly reduces the variability of the aerosol number concentration and thereby increases stationarity, has been added to section 3.3. We agree that the word advection was not a good choice of word to use throughout the manuscript. We have changed the term LIA (Low Influence of Advection) to LAM (Low Influence of Anthropogenic sources and Mineral dust). In the figure text under Fig. 10, we found an error. In Fig. 10a, also half-hours that do not fulfill LAM conditions have been included. This was not stated in the figure text before but is now. (However, data collected during rainfall and the wind sector associated with risk of pollution from the diesel generator is excluded). We agree that the half-hours with the largest variations in particle concentration in Fig. 10a are not even close to fulfilling stationary conditions. However, in this figure we are most interested in showing how the sign of the flux is affected by decreasing concentration and not so much of the magnitude of the flux. Half-hours with the largest decreases in concentration in Fig. 10a have been affected by pollution before entrainment results in decreased particle concentration. These cases are excluded in the data filtering process in section 3.3, and thereby not affecting the particle transfer velocities in Eq. 8 or the diurnal cycles of the particle fluxes in Fig. 9.

Referee comment 4:

Equations 1, 4, 5, and 6 are garbled on the screen and when printed and must be redone in that they appear to be incorrect in their scrambled format available through the review process. One can guess that the authors used the correct equations through the way that the equations are garbled (somehow).

Response:

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The equations were garbled in the first manuscript we sent in. However, we made some technical corrections before the manuscript was published on the ACPD website. The equations look fine on our screens and also when printed, for the latest version. We do not know whether the reviewer might not look at the latest version of the manuscript we sent in. We leave this matter for the final technical check by the ACP staff, but don't expect it to be a problem.

Referee comment 5:

Lines 83-84: be careful with wording here, entrainment was shown to be important not emission

Response:

We have changed this statement and added the possibility of entrainment for producing upward fluxes. We have also changed "would" to "could" both in this sentence and the following one.

Referee comment 6:

Line 94: "contribute"

Response:

We have changed this.

Referee comment 7:

Line 126: add units for LAI.

Response:

We have added $m^2 m^{-2}$.

Referee comment 8:

Lines 294-295 how were equations 1-2 used; were the corrections ever applied to the

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final data?

Response:

As was stated on lines 218-219, all fluxes in this study have been corrected according to Eq. 1-2.

Referee comment 9:

Lines 296-302: what did co-spectra of heat and vapor fluxes show? Were they similar?

Response:

The spectra of heat and water vapor generally looked as they should considering similarity relationships. We had to make some limitations on how many spectra to show and thought it was a good idea to at least show flux spectra that involves all instruments. The sensible heat flux spectra involve the same instrument as the momentum flux spectra, and the water vapor flux spectra involve the same instrument as the CO₂ flux spectra. We have added this information to the manuscript.

Referee comment 10:

Line 377: the friction velocity is actually > 3 times as high in the day, not "slight".

Response:

"slight" has been removed.

Referee comment 11:

Line 399: give r² for this "correlation".

Response:

We are not sure what equation this comment refers to. At line 399, studies by other authors are discussed concerning how much biogenic organic compounds may contribute to the mass concentration of BC_e. If comment 11 refers to Fig. 7a, the r² value

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is 0.31. However, the point with this figure was that the aerosol number concentration increases with increasing B_{Ce} when B_{Ce} is high (roughly for B_{Ce}>80 ng m⁻³. For lower B_{Ce} concentrations, the aerosol number concentration is more or less constant. Thus, it seems that anthropogenic particles dominate the aerosol number population when concentrations are high but have a much lower impact when concentrations are low. We have rephrased the text in this section to clarify what we mean. We also have added 25 and 75 percentiles to Fig. 7b to provide some information of the uncertainty.

Referee comment 12:

Line 439: processes may be more numerous but not necessarily "More complex"; see lines 664 443 (other errors) are more difficult to quantify...but counting error dominates"? You give no citation nor analysis to support this. What about hygroscopic growth?

Response:

The words “more complex” have been removed from the manuscript. Even though the aerosol number concentration in the Amazon boundary layer is relatively stable in the wet season (compared to aerosol concentration in most other areas), it is much less stable than the CO₂ concentration. The higher day to day variability in aerosol concentration compared to the day to day variability in CO₂ concentration is likely the most important factor for the larger variability in aerosol flux compared to the CO₂ flux. Also, the fact that entrainment may produce upward fluxes on days when entrainment has a lowering impact on the aerosol number concentration within the boundary layer of course increases the day to day variability. Larger uncertainties in the aerosol flux measurements compared to the CO₂ flux measurements may play a role as well. The sentence “However, the uncertainty due to discrete counting is most likely the major source of errors in the aerosol flux” has been removed. We expect hygroscopic growth to have a minor influence. The reason is the very low numbers of nucleation mode particles in the Amazon boundary layer in the wet season (Zhou et al., 2002; Rissler

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et al., 2004). Also, the average diameter growth factor of particles in the wet season is relatively low, 1.3 taken from dry to 90 % relative humidity (Rissler et al., 2004).

Referee comment 13:

Lines 570-574 Also higher background reduces new particle nucleation compared to coating the existing particles with any new secondary organic material (due to surface area).

Response:

Still, new particle formation has been observed only occasionally in the boundary layer over the Amazon rain forest and it shows very different patterns from typical nucleation events known from the mid-latitudes, for example. In the Amazon Basin the smallest particles are typically 10-20 nm (Martin et al., 2008; Zhou et al., 2002), which has lead to hypothesis that new particle formation may takes place at higher altitudes in the free troposphere and/or in association with convective clouds (Ekman et al., 2008; Krejci et al., 2003).

Referee comment 14:

Line 581: Held et al. 2006 citation is not in the reference list.

Response:

This reference has been added.

Referee comment 15:

Lines 622-624: Excluding periods with likely HIGHER anthropogenic influence does not mean that all anthropogenic influence is gone.

Response:

This is also explained, that there still may be some anthropogenic influence on our data set even after the data filtering process. This is the reason why we make the

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investigation in Fig. 14 of how the sign of the flux depends on B_{Ce} concentration (and also on aerosol number concentration in Fig. 14a).

Referee comment 16:

Furthermore, many secondary particles trace their origin back to anthropogenic emissions like SO₂. This distinction is poorly made in this paper, if at all.

Response:

Yes, anthropogenic SO₂ from primarily Manaus could play an important role in new particle formation. However, it must be stated that Manaus is located 60 km SSE of the measurement tower and the very dominant trade wind direction was between E and NE. So, even if some influence from Manaus survived our data filtering process, these data will not dominate the median particle flux since these events will be too few in numbers.

Referee comment 17:

Lines 629-30: There is no way to separate deposition of natural from anthropogenic particles in this study as it is presented. Too many nearby anthropogenic emissions present a problem but what difference would there be between deposition of long range transport of natural or anthropogenic emissions (unless receptor modeling and chemical tracers could be somehow utilized)?

Response:

As was described in sections 2.3.2 and 3.3, we have used a chemical tracer for anthropogenic influence on the aerosol population. Mass concentration of equivalent black carbon (B_{Ce}) was used as a chemical tracer using a Multi-Angle Absorption Photometer (MAAP). We have also done a careful filtering of data by using B_{Ce}, wind direction and aerosol number concentration. Fig. 14 illustrates how the particle transfer velocity is affected when going from higher B_{Ce} concentrations to extremely clean conditions. There it can be seen that the flux points downward even in the absolute cleanest con-

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ditions.

Referee comment 18:

Line 688: A different equation is presented for V_d vs U^* here than in the abstract or text. Is an intercept needed or not? what is the R^2 for this equation in either form?

Response:

We have added an intercept in this equation in the summary and conclusion section. The correlation coefficient was given straight after Eq. 8.

References not found in the manuscript:

Andreae, M. O.: Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions, *Atmos. Chem. Phys.*, 9, 543-556, 2009.

Araújo, A. C., Kruijt, B., Nobre, A. D., Dolman, A. J., Maarten, J. W., Moors, E.J., and Souza, J. S.: Nocturnal accumulation of CO_2 underneath a tropical forest canopy along a topographical gradient, *Ecological Applications*, 18(6), 1406-1419, 2008.

Artaxo, P, and Maenhaut, W.: Trace element concentrations and size distribution of biogenic aerosols from the Amazon basin during the wet season, *Nuclear Instruments and Methods in Physics Research*, B49, 366-371, 1990.

Marcolla, B., Cescatti, A., Montagnani, L., Manca, G., Kerschbaumer, G., and Minerbi, S.: Importance of advection in the atmospheric CO_2 exchanges of an alpine forest, *Agric. For. Meteorol.*, 130, 193– 206, 2005.

Roberts, G. C., Andreae, M.O., Zhou, J., and Artaxo, P.: Cloud condensation nuclei in the Amazon Basin: "Marine" conditions over a continent?, *Geophysical Research Letters*, 28, 2807-2810, 2001.

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