We are very grateful to the reviewers for their comments and suggestions. The manuscript has been substantially reformatted and modified based on these suggestions and the conclusions have been amended. Detailed responses to each of the review's comments are given below. All equation, section, and figure numbers in the responses below refer to the previously submitted manuscript, not the newly revised manuscript. Reviewer comments are in black, and the responses to the reviewer comments are in blue.

Referee #1

While the number of aerosol flux measurements by micrometeorological techniques is still very limited, the situation is even worse for size-resolved fluxes and deposition rates. The paper brings important new data and insight to the problem applying a fast mobility particle sizer combined with the eddy covariance technique (EC). The new results are also utilized in the interpretation of earlier observations at the same location by a quadrupole aerosol mass spectrometer providing chemically speciated information.

Major:
1. p. 22481, lines 1-8: Why is it especially the incoming solar shortwave radiation. which has adopted as a explaining factor? The role of SW is explained and discussed very superfluously.
SW radiation is only used as a proxy for the peak in photochemical activity. SW radiation itself is not responsibly for anything, but the peak in photochemical activity can be correlated to sulfuric acid nucleation, particularly when significant SO2 is present. We have added some text explaining this and relevant references for nucleation and growth which have been studied extensively.

2. Section 5.2, Effect of precipitation: What does the leaf wetness exactly mean and what are the values the sensor is providing? What does mean the leaf wetness of 10%? 10% of what? Namely, some/many leaf wetness sensors provide values which are mainly on/off, depending whether the surfaces are dry (although possibly having very thin water film) or wet and not really anything between. I raised up this issue also since it is speculated that the aerosol deposition could be larger on wet surfaces, but no convincing physical explanation is given for that. I am surprised on the statement "there are a small number of flux measurements during rain". I do not believe that anyone can measure reliably fluxes by EC during the (heavy) rain.
(a) Description of the sensors has been added to the Instrumentation section. The leaf wetness sensors measure the presence of water on the sensors (which mimic the leaves). 10% is an arbitrary reading and the output of the sensor has been corrected to a binary wet/dry reading.
(b) At this time we can offer no convincing physical explanation for the larger deposition to wet surfaces.
(c) While it is true that many open path IRGA instruments do not function well during rainfall (e.g. Dias et al., 2009), sonic anemometers can function without much data loss (c.f. Grellea, 1997) if the rain is not intense. During this study, there was no heavy or intense rain and analysis shows no increase in rejected data during rainfall.

3. p. 22483, lines 15-16: I do not understand the meaning of "..that either the positive flux is balanced by a negative flux..."

This sentence only reiterates what was said earlier in the paragraph. Since it is confusing and unnecessary, it has been removed.

4. Section 5.5. Decoupling of the canopy space: Instead of using the friction velocity as a criterion for the decoupling, the more appropriate factor could be the canopy Richardson number (see Mammarella et al., Determining the contribution of vertical advection to the net ecosystem exchange at Hyytiälä forest (Finland). Tellus 59B, 900-909, 2007). More importantly, I am very surprised that the time lag during low friction velocity conditions is order of hours. This is very slow and normally the mixing even under low-mixing conditions occurs on the time scales of the order of 100 seconds. If the time lag is real, the explanation must be something else that the low mixing, but I am also wondering how so long time lag can exist if the friction velocities are 0.1 m/s or larger, since although they are low they should induce some mixing and thus the conditions are not very stable. There is something strange in the decoupling result and/or its interpretation.

The friction velocity is replaced in the figure and discussion with the Richardson number, as defined in Mammarella et al. (2007). The frictions velocities of 0.1 m/s are above the canopy. Friction velocity measurements made during the campaign at a 2m height (not included in the manuscript) were near 0.03 m/s at night. Hence mixing would be much slower beneath the canopy. Although we agree that the decoupling seems intuitively strange, a similar effect was also seen in a recent study by Whitehead et al. (2010, their fig 10), with night-time delays "as much as 3 hours". Discussion of the Whitehead et al. study has been added to the manuscript.

5. I am not sure that the sections 5.6 Speciated results and 5.7 Amonnium-nitrate evaporation are needed in this paper. They are related to the earlier measurements and does not bring very much insight to the actual topic of this paper. Instead, the section 5.8 has deserved its place. If 5.6 and 5.7 are omitted, it must be checked that 5.8 is still understandable as such.

Following the comments of the second reviewer (major comment #1), these sections are retained but modified to further investigate the possible effect of ammonium-nitrate evaporation on the flux direction.

Minor:

1. Eq. 1: there is a mistake, the advection term should be $u_i \frac{\partial C}{\partial x_i}$. Similarly to the term including V_g.

In the original conservation equation (e.g. Businger, 1986), the velocities are within the derivative. It is moved outside, after the assumption of incompressible flow, which has been added to our list of assumptions.

Explain what is x_i and u_i and the summation notation over i indices.

Added text: "and the subscript i is a summation index such that $(x_1, x_2, x_3) = (x, y, z)$ and $(u_1, u_2, u_3) = (u, v, w)$."

I would not call C as a scalar concentration but as aerosol particle concentration since it is implicitly assumed that the scalar relates aerosols since the equation includes the gravitational settling velocity, although it is assumed to be negligible later. Similarly, replace the concept of “molecular diffusion coefficient” to “particle diffusion coefficient” or “to diffusion coefficient due to Brownian motion”.
Changed.

2. Eq. 4: define D and D_0. Units of RH is %, but now it seems that RH has values between 0 and 1, and it is rather the saturation ratio and not the relative humidity.
3. p. 22476, line 17: related to the comment above, RH is the saturation ratio. I guess q is not the specific humidity ratio but specific humidity.
The definitions of D and D_0 are added. Percent is not a unit; it is equivalent to 1/100. To avoid confusion, RH is replaced with saturation ratio ($0< S_R < 1$), definitions are added, and q is removed.

4. p. 22477, lines 1-2: I do not understand the sentence, what is the average increase over all size ranges and what is a total decrease, and how you have ended up to the results (26% and 5.8%)?
The sentence is rewritten as “The average corrections can be represented as the average ratio of corrected to uncorrected fluxes ($<F/F_0>$) or as the ratio of the average corrected flux to the average uncorrected flux ($<F>/<F_0>$).”

5. p. 22494, line 32: replace “Lilavainen” by “Lihavainen”
6. p. 22495, lines 15-16: replace “Launianinen” by “Launiainen”
7. p. 22495, line 31: replace “Vesela” by “Vesala”
Changed.

8. Fig. 1: I cannot read what is the scale.
9. Title fonts are much too small in Figs. 2, 3, 7, 11 and 13.
The scale and fonts of all the figures have been changed to improved readability.

10. Fig. 3: Is the data averaged over the whole measurement period? If yes, it is so called averaged daily course. The caption does not mention plot a).
The term “diurnal variation” is a more common expression than “averaged daily course”, and the data are presented as percentiles, not averages. “Number of 30-min measurements” was mentioned out of order. It has been moved to the start of the caption.

11. Fig. 11: explain in the caption what is the light blue curve.
Explanation added.

Referee #2

The manuscript by Gordon et al. presents size-segregated aerosol flux measurements above a Canadian mixed forest, made with a relatively new fast (1 Hz) particle spectrometer (FMPS). It also contains some of the first chemically resolved aerosol flux measurements by eddy-covariance, using an Aerosol Mass Spectrometer (AMS). The current understanding of the dry deposition process of particles remains incomplete with large differences between models and measurements and unexplained variability between sites and studies. As such the measurements make a useful contribution to the literature, especially because the study attempts to link size-segregated observations with chemically speciated observations, albeit from measurements conducted during different summers. The data analysis is mainly sound and the English is clear. However, there are a number of major scientific concerns that need to be addressed before the paper can be considered for publication in ACP.

Major Comments:

(1) In my opinion the authors miss the most likely explanation for upward fluxes. Nemitz and Sutton (2004) refer to several studies that have observed simultaneous apparent (measured at the reference height) emission of small particles, while larger particles showed deposition. They continue to show that this behaviour is consistent with the effect of ammonium nitrate (AN) evaporation during the deposition process, if fluxes are measured with an instrument with fixed bin sizes. Because particles undergo shrinkage during the deposition process, fluxes are derived according to a parameter (i.e. diameter) that is no longer conserved with height. This is the same effect as caused by water loss due to RH gradients discussed by the authors. The earlier AMS flux measurements at this site appear to confirm that deposition velocities were much larger for nitrate than for sulphate, a fact that suggests that AN evaporation was affecting chemical bulk fluxes. Thus it is also highly likely that this process was also affecting particle number fluxes. As such the information on the sulphate / nitrate exchange is in my opinion central to understanding the particle number fluxes, and rather than omitting Sections 5.6 and 5.7 as suggested by Reviewer 1, I suggest to fully integrate the AMS measurements into the paper, with more information on the methodology given in the methods section. I believe that after these earlier studies have been considered, the conclusions of the paper will change significantly.

(a) Although we observe emission of smaller particles, it is not true that larger particles show deposition. Even when fluxes are very small for large sizes, the median values are always positive. If the number fluxes are used to derive volume fluxes, the average size-segregated volume flux consistently increases with bin size over the full range of the instrument. We are hesitant to include this in the manuscript, as the larger size measurements (even when binned together) are below the FMPS noise level and outside the calibrated size range (as shown in Fig. 2).

(b) We agree with the reviewer's comments and have added a section discussing the evaporation of AN. In this section the false fluxes caused by AN evaporation are investigated following a derivation similar to the deliquescence correction. It is shown that, although a correction due to AN evaporation may change the flux direction at small particle sizes (*D* < 70 nm), above this size AN evaporation does not likely explain the apparent upward fluxes. Similarly, Nemitz and Sutton (2004) consider four model scenarios and observe a false upward fluxes in two scenarios (for *D* < 300 nm and *D* < 100 nm). Even in these scenarios, false downward fluxes are seen for larger particles, which would result in an increased positive mass flux in our study.

(2) Overall, the structure of the paper is unusual in that the Discussions Section accounts for 9 Figures out of a total of 15. As a result a lot of experimental detail is introduced only very briefly where measurements are referred to or it is missing altogether. All experimental details should be moved into the Method section, not only on the AMS flux approach, but also on the SMPS, the leaf wetness measurements (what kind of sensor was used?), the NH3 and SO2 measurements etc. The basic flux, AMS and SMPS measurements (Figs. 11, 13 & 15) should be shown in the Results section and only figures that refer to the interpretation of the results (e.g. Fig 14) should remain in the Discussion Section.

The manuscript has been reformatted as suggested. Both studies (2006 and 2009) are now discussed in parallel. Figures 13 and 15 are moved, but Fig. 11 is left in the discussion section, as it follows from the investigation of measured flux and storage flux.

(3) The authors present fluxes for individual particle sizes throughout the manuscript, which is not meaningful. For example, C62nm and F62nm are probably zero as there are no particles with a diameter of exactly 62 nm. Instead, the measured fluxes actually represent the flux within a size-range as given by the size bins of the FMPS. Therefore, throughout the paper, any quantification of a flux needs to be accompanied by the width (or better range) of the size bin to which it refers, or it needs to be normalised by the bin width, i.e. presented as dC/dlogDp or dF/dlogDp (cf Fig. 4c; Ahlm et al., 2010). For the same reason, Fig. 2 should be presented as dC/dlogDp in order to make it more generic and independent of the bin width of this particular instrument, and Fig 4a should be represented as dF/dlogDp. This is not applicable to exchange velocities where the width of the bin cancels out when dividing the flux by the concentration.

References to a single size have been changed to a range throughout the manuscript. The ranges also now incorporated the minimum and maximum diameter of the bin size ranges (e.g. 18 < *D* < 452 nm as opposed to 20 < *D* < 410 nm, where 20 and 410 nm are the median sizes of the bins). Data are now presented as dC/dlogDp and dF/dlogDp in Figures 2, 4, 6, and 7.

(4) It remains unclear, what corrections were applied to the data for which part of the analysis and what the rationale was. For example, were the data corrected for the effect of RH fluxes? Were fluxes corrected for storage error (Section 4.3) prior to further analysis (e.g. for Table 1)? The authors should more stringently distinguish between (i) fluxes as measured at the measurement height, (ii) best-estimate fluxes at the measurement height, corrected for hygroscopicity (if required, depending on whether the FMPS measures dry or wet size, see below) and the effect of NH4NO3 volatilisation on size (see above) and (iii) the best estimate of the actual surface / atmosphere exchange (corrected for chemistry and storage between vegetation and measurement height). Although the authors may not be able to quantify all these effects explicitly, these issues should

nevertheless be discussed with these differences in mind. The authors should make it more explicit where they are talking about the (uncorrected) fluxes measured at the measurement height. For example, the upward fluxes are only ‘apparent’ upward fluxes, they are unlikely to be emissions of particles from the vegetation surface. It is also not completely clear for which analyses the data were filtered, e.g. for low u* and for which they were not. This has implications for the interpretation of the results (see below).
The data were corrected for variation in density due to the RH fluxes according to the Webb correction, as stated in Section 3.1 (para 2) and hygroscopicity (also stated in Sect. 3.1). We disagree that the storage rate (term 1 in Eq. 3) should be considered an error to be corrected; rather it is one component of the budget that can be quantified separately. Equation 3 demonstrates the relationship between measured flux, storage rate, and the net sum of sources and sinks. Section 3.1 has been modified to make the application and extent of the corrections more clear. Changes have also been made throughout the manuscript to highlight differences between measured fluxes at 33m and estimated surface/atmosphere exchange, including the following text: “From herein all presentation and discussion of flux (*F*) refers to the corrected flux measurement. It is emphasized that these apparent flux measurements may not be equal to the exchange of aerosols between the canopy and the measurement height above (S_T), due to storage (Eq. 3) and unresolved chemical effects, which are discussed in Section 5.”

(5) I find the discussion of the effect of particle growth on fluxes (Sections 5.1, 5.7 and Conclusions) confusing as it appears to combine horizontal and vertical transport phenomena. The particle growth detected by the SMPS is a regional scale phenomenon that presumably occurs over large areas. Thus, changes at the measurement height are more likely to reflect processes that occur along horizontal gradients rather than provide information on in-canopy processes below the measurement height. The main effect of the horizontal changes on fluxes would be through the storage error which the authors quantify separately. This has important implications; for example, the decrease in nitrate (p22488, l4) is likely to have been due to an increase in temperature overall, shifting the equilibrium from the aerosol towards the gas phase. Rather than being driven by vertical gradients this is again a regional phenomenon. In addition, temperature is only one factor affecting the nitrate partitioning and its change with height, the others being gradients in RH and, importantly, the removal of NH3 and HNO3 by the canopy, which results in very small gas-phase concentrations near and within the canopy. Without simultaneous flux measurements of NH3 and HNO3, the effect cannot fully be quantified. Further, the vapour pressures of NH3 and HNO3 also respond to other aerosol components such as sulphate.
We agree that the changes in concentration are better explained by regional phenomena. We have removed comparison of the NO3, NH4 concentration and the NO3 flux (including Fig. 14), and have instead limited our discussion to the connection between vertical temperature differences and NO3 flux.

(6) Linked to this, the discussion of the effect of NH4NO3 evaporation and the attempt to quantify the effect is incomplete. The authors imply that the additional flux induced by the evaporation is constrained by the increased in NH3 storage within the canopy. This argument misses two facts: firstly, as the authors concede later in the text, other factors

are much more likely to dominate the variability in the NH3 concentration, such as the gas/aerosol partitioning at the regional scale, changes in the competing HNO3 concentration (not measured), changes in boundary layer height and, most basically, changes in air masses that are advected to the measurement site. Secondly, and more importantly, the argument ignores that an increase in the deposition rate also increases the NH3 deposition flux and thus NH4NO3 volatilisation can increase the effective (or apparent) NO3 deposition rate without increasing the NH3 concentration. For example, van Oss et al. (1998) modelled the effect of NH4NO3 volatilisation on bulk chemical fluxes, based on the data of Wyers and Duyzer (1997).
These dynamics are complex and it is difficult to draw conclusions without much more detailed measurements. For these reasons, discussion of the relationship between NO3 flux and NH3 storage (Fig. 14) has been removed from the manuscript.

(7) In the present analysis there is disagreement between the total mass fluxes derived from the FMPS fluxes and those derived from the AMS. The possibility of some organic fluxes not having been resolved by the AMS provides a convenient explanation. However, in my mind it is much more likely that if the FMPS fluxes were properly corrected for the effects of storage (it is unclear whether this correction was applied, see above) and changes in size due to NH4NO3 volatilisation, these difference may be resolved.
As discussed above, storage is not considered a correction (see Eq. 3). It is discussed in Section 3 that, when averaged over a time period of many days or weeks, the storage term should tend to zero. Hence, it is unlikely that the effects of storage will change the comparison between studies. There may also be diurnal biases in the number of samples per hour, which is discussed. NH4NO3 volatilisation is also discussed as a possibility, since the FMPS is a size resolved instrument and the fluxes will be affected by the size change (as pointed out by the reviewer in the first comment). However, the reason for the difference remains unresolved.

Minor Scientific Comments:

Abstract. The abstract talks about a “net production rate” of 75 nm particles. At no point does the manuscript claim that 75 nm particles are produced (indeed, there is unlikely to be a mechanism for this). In addition, it does not make sense to quantify the flux of a size for the reasons stated above. Thus, the sentence should use wording such as “the size-distribution of the net apparent upward flux at the measurement height peaked at a particle size of 75 nm”.
The sentence is changed to: “The size distribution of the apparent production rate of particles at 33 m peaked at 75 nm.” As discussed above, we wish to distinguish between apparent flux, storage rate, and production rate (= flux + storage, from Eq. 3). A possible mechanism for the peak at 75 nm is that the growth rate of the particles at that size is slower than it is at smaller sizes. This is added to the text.

P22471, l1. This sentence is somewhat imprecise. SO4= cannot lead to eutrophication only to acidficiation, but NH4+ can also contribute to eutrophication.
Changed.

P22472, l15. The historical summary of aerosol flux measurements is incomplete. There are other eddy-covariance datasets of size-segregated particle number fluxes, many of which are summarised in the review of Pryor et al. (2008b). Key datasets include those of Gallagher et al. (1997) and Ahlm et al. (2010). Overall, the application of optical particle counters to the measurement of size-segregated particle number fluxes predates the application of CPCs to the measurement of total number fluxes, e.g. Sievering (1987), although some of the earliest measurements were not made over forest.
We are restricting the discussion here to size-segregated, sub-micron particle number fluxes over forests. The omission of Gallagher et al. was an oversight, and that paper has been included. Hicks et al. 1989, in which upward fluxes of small particles were measured, was also overlooked and is included. Ahlm et al. 2010 is also added.

P22473, l14. Although the FMPS reports data at 1 Hz, was this the actual response time of the system (inlet + analyser)? Has this been validated by inducing step changes in concentration and looking at the response?
The use of the term 'response time' is confusing, as it seems to imply a time lag, which would have no effect of the instrument frequency. We have modified the text to refer to either the frequency response (assumed 1 Hz) and the time lag. A step change test would presumably give both the lag (the delay between the introduction of the step and the measurement of the step) and the frequency response (the extent that the step is 'smoothed' in the measurement). Although we have not done this test, the response correction is not very sensitive to the instrument frequency. Halving the frequency to 0.5 Hz results in an average change in the corrected flux of < 5%.

P22473, l19. What was the flow rate of the FMPS through the 1/4" inlet? What was the Reynolds number?
The flow rate through the inlet is 10 L min^{-1}. The reported residence time of 0.8 s was miscalculated and has been corrected to 0.3 s. This gives a Reynolds number of approximately 4000. This has been added to the text.

P22474, l5. If for the larger sizes, concentrations from several bins were bulked together, it may be possible to derive fluxes for larger sizes. This may show the bidirectional flux behaviour with simultaneous apparent emission of the smaller sizes at times when the larger particles show emission, as would be expected for NH4NO3 evaporation.
As discussed in the response to major comment (1) above, this effect is not seen (even when the bins are bulked together).

P22477, l1. To what extent are particles dried during sampling by the FMPS? Was dry sheath air used? The answer to this question determines whether the hygroscopicity correction is applicable or not. Related to this, Khlystov et al. (1996, 1997) noted that NH4NO3 evaporation may occur in DMPS/SMPS systems. Are FMPS systems likely to be affected also? What would be the effect on the measurement?
The following text is added: "Since the FMPS mixes sample air (8 L min^{-1}) with temperature controlled sheath air (40 L min^{-1} at 23^{o}C), the extent to which hydroscopic particles are dried in the FMPS sampling chamber is unknown. In the following analysis,

we assume there is minimal drying and the deliquescence correction is applied to the fluxes. However, the uncertainty in the flux measurements due to this unknown is equal to the correction, which is smaller than 6% on average." Further, flow rates in the FMPS are much higher than in the DMPS/SMPS systems, so there should be less NH4NO3 evaporation. However, this is also very difficult to quantify.

P22477, l21. How can the size range around 62 nm (again this should refer to the bin, rather than a single size) be a good representation for the total size range if it reflects the peak (and thus an extreme) in the size distribution? Why not calculate the total flux over the range and use this for most of the analysis?
This was poorly worded and has been rewritten to say that the exchange velocities are similar. We have changed the analysis to look at total flux in Sections 5.3 and 5.4 (Figs. 9 and 10). However, we have kept the analysis of the 62 nm size bin fluxes ($57 < D < 69$ nm) in other sections. Since we are investigating the cause of the apparent upward fluxes, it makes sense to look at the bin size with the largest upward fluxes.

Section 5. The authors may want to consider relating their work to the observations of Whitehead et al. (2010) over South-East Asian tropical rainforest who present a similar time-scale analysis of decoupling.
We are very grateful to the reviewer for bringing this study to our attention. The Whitehead et al. results have been included in the manuscript.

Section 5.3, Fig. 10 & Eq. 11. What is the effect of filtering for low turbulence on Eq. 11? Presumably FStg is not equally distributed over day and nighttime conditions, and thus periods of high and low turbulence.
The analysis was done with and without filtering for friction velocity, with no significant effect on the results. A discussion of this testing is added to the text.

P22486, l22. In the expanded method section (see above), the authors need to clarify that the AMS flux mode is distinct from the standard AMS JMS mode in that it keeps the beam open for 28 minutes.
Text added.

P22486, l27 & Fig. 15. Presumably m/z 43 was not the only organic fragment monitored by the AMS? Were the fluxes of the other fragments consistent with the behaviour of m/z 43? Could they be integrated into the analysis to derive more robust total organic fluxes?
The fragments m/z 44 and 57 were also measured; however they showed weaker correlation (r2 = 0.77) with the total organic aerosol mass (measured in the MS mode). Further, the m/z 57 signal was very weak (ca. a factor of 0.1 times the m/z 43 signal) and not considered reliable. Hence these data were not included in the analysis.

Technical Comments

P22470, l20: ". . . range from a few nm to tens of"
P22475, l18: " . . . used to rotate the anemometer measurements . . ."

P22475, l25: “. . . of the signal carried by frequencies > 1 Hz due to . . .”
P22476, l12: “Swietlicki”
P22478, l10: “Rannik et al. (2009) demonstrated that . . .”
Changes made.

Additional references (not in manuscript or reviewers’ comments)

Dias, L.N., Hong, J., Leclec, M.Y., Black, T.A., Nesic, Z., and Krishnan, P.: A simple method of estimating scalar fluxes over forests, Boundary-Layer Meterol., 132, 401–414, 2009.

Grellea, A., Lundbergb, A. Lindrotha, A., More´na, A.-S., Ciencialaa, E.: Evaporation components of a boreal forest: variations during the growing season, J. Hydrol., 197, 70–87, 1997.