Interactive comment on *"Ammonia emission fluxes for beech forest after litter fall – measurements and modelling"* by K. Hansen et al.

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Authors' response to Anonymous Referee #2

We thank Referee#2 for valuable comments and constructive suggestions. We believe that taking into account these comments and suggestions have led to a considerable improvement of the paper. In the following we go through the comments by the reviewer point by point, and explain the steps taken in the revision of the paper:

Referee comment #1: *This manuscript presents important data on NH_3 fluxes above a senescing forest measured by relaxed eddy covariance. The NH_3 concentration from a regional NH_3 model is also compared to measured concentrations, comforting the hypothesis of local NH_3 emissions leading to increased NH_3 concentration. The authors suggest that the emission fluxes observed following leaf fall were due to both possibly increasing NH_3 emissions from leaf litter and diminishing NH_3 emissions from leaves.*

However the authors do not give substantial elements allowing to weigh which process may be dominant (increase in litter emissions and/or decrease in leaf absorption). The increased emissions may indeed be due to a combination of increased litter emissions, decreased (zero) leaf absorption, but also decreased turbulent transfer in the canopy with a decreased LAI. The authors should discuss further (and also provide a graph) the change in canopy turbulent resistance induced by the change in LAI. The manuscript anyway reports very rare data - on NH_3 fluxes over a defoliating forest - which are essential to publish. I would therefore recommend publication of the manuscript.

Response: First of all, we are very pleased that the reviewer finds the paper interesting and recommend publication. We measure the ammonia flux above the forest in the autumn 2010 in order to assess ammonia emission fluxes related to leaf senescence as flux studies in such periods are rare. We compare our results to a state-of-the-art local-scale Gaussian dispersion-deposition

model (OML-DEP) to assess the magnitude of the contribution of the emission fluxes from the forest. As the data set of NH_3 flux measurements is relatively small and represents a period characterized by large variations in forest's phenology, meteorological and soil conditions that influence the atmosphere-forest exchange of NH_3 it is not reasonable to weight which process that may dominate the atmosphere-forest exchange of NH_3 (increase in litter emissions and/or decrease in leaf absorption). However, we understand the reviewer's point that decreased turbulent transfer in the canopy during the leaf fall period could influence the NH_3 flux, and we are grateful that the reviewer suggests us to investigate look into the canopy turbulence using the resistance terms. However, as the resistance terms are parameterisations more than actual physical quantity, we chose to calculate the canopy turbulent resistance using the measured u_* that indirectly represents the LAI/PAI of the canopy. We calculated the turbulence intensity as u* normalized by the wind velocity $TI = \frac{u_*}{U}$ and added this to paragraph "2.3 Local meteorological measurements". We did not see any relationship of between PAI and the turbulence intensity following like decreased turbulent transfer (increased resistance) due to the leaf fall. However, we found that highest intensities were related to the short fetches when wind came from easterly and westerly directions (Fig. 5a) and this could have contributed to enhance the emission fluxes during the short episodes with emission peaks. This has been included in the discussion in paragraph "4.1 LAI and NH_3 fluxes in the leaf fall period" along with a figure showing the turbulence intensity (Fig. 6A).

Furthermore, we believe that title of the manuscript have caused wrong expectations to the analysis of this study. In order to make the title of this manuscript illustrating the content more precisely, we have therefore changed the title to "Ammonia emission fluxes for deciduous forest after litter fall".

Referee comment #2: *Section 2.2. I wandered whether these LAI measurements should take account of alometric distributions? Any comment on that?*

Response: The LAI measurements performed using the LAI 2000 does not take the alometric distribution of the trees into account. As this study has its focus on the ecosystem-atmosphere exchange processes we use LAI to represent the canopy development and we believe that this is reasonable and adequate and for the purpose of this study.

Referee comment #3: *Page 5 Line 2: Could the authors be more detailed expansive on the material used for meteorological measurements?*

Response: The climate and environmental parameters that was obtained from the European Fluxes Database Cluster is explained in a paper by Pilegaard et al. (2011). The paragraph "2.3 Local Meteorological measurements" has been changed to:

"The wind components in x-, y- and z-directions were measured at 10 Hz sampling using an ultra-sonic anemometer (Metek-uSonic-3 Scientific) installed above the forest canopy at 34 m height. Half-hourly averaged values of wind velocity, wind direction, friction velocity, temperature, and Monin Obukhov Length were conducted from the 10 Hz sampling and the turbulence intensity was calculated as the friction velocity normalized by the wind velocity as $(\frac{u_*}{U})$. Precipitation, relative humidity, soil temperature, soil water content, and global radiation were obtained from the European Fluxes Database Cluster (www.europe-fluxdata.eu) and are described in Pilegaard et al. (2011)."

Referee comment #4: *Page 5 Line 10: What is a "micrometeorological relationship"? This is very vague. The authors should be more precise here.*

Response: The sentence has been specified to:
"REA is combining measurements of the vertical momentum flux and the difference between the average trace gas concentration of upward and downward moving eddies."

Referee comment #5: *Page 5 Line 20: The "proxy scalar" is heat (ïA¸šCpT), please precise.*

Response: The sentence has been specified to:
"However, turbulent transport, especially over very rough surfaces, often violates the underlying assumption of a linear relationship between w and C (Ruppert et al. 2006), thus the use of a b-coefficient determined from a proxy scalar (i.e. the sensible heat flux (H)) better reflects the correct b-coefficient for a certain measurement period."

Referee comment #6: *Page 5 eq. (3) and Page 6 Line 1: Please rather use H for the heat flux symbol as it a more common symbol.*

Response: Tflux has been replaced by H throughout the manuscript.

Referee comment #7: *Page 6 Line 5. Why choosing b0= 0.6 when the range is 0.4-0.6? 0.5 would have been more logical. Please justify.*

Response: The interval 0.4-0.6 is found from a number of studies in the literature, but we chose to use the 0.6, following the study of Oncley et al. (1993).

Referee comment #8: *Page 6: Lines 9-11: Does the height difference between the sonic and the WEDDs make a problem? Please comment on this.*

Response: We believe that the 10 l/min air flow into the inlet causes a flow distortion around the inlet that could affect the air samples. According to a study by Kristensen et al. (1997) the loss of fluxes is less when placing the scalar sensor under the anemometer than when sensor separation is longitudinal. This was the main reason to deploy the sensors as described in the paper.

Referee comment #9: *Page 6: Line 21: Is it NH3 or NH4+?*

Response: It is NH_4^+ and has been corrected in the revised manuscript.

Referee comment #10: *Page 6: Line 30: Was the symbol CRNH3 defined already?*

Response: We have now defined the symbol c_{RNH3} earlier in the paragraph.

Referee comment #11: *Section 2.5: A mention to Martin Ferm would be appropriate in this section as he is the "father" of NH3 denuders.*

Response: A reference to Ferm 1979 has been included in section 2.5 "Denuder measurements".

Referee comment #12: *Page 7: Lines 13-16: I do not follow the reasoning there. Please rephrase and explain what you are meaning.*

Response: As we are comparing measurements of the atmospheric NH_3 concentration above the forest from two different systems deployed in two different measurement heights, the measurements may not be directly comparable. However, the difference between the mean concentration measured in the period 26 October – 11 November 2010 using denuders (29.8 m) and the REA method (33 m) was only 0.01 µg NH_3-N m^{-3} indicating that the measurements are comparable. Furthermore, as the uncertainty of the REA measurements of NH_3 concentration is relatively large we expect the denuder NH_3 concentration c_{dNH3} to be within the uncertainty interval of the REA measurements. We have removed the argument using Andersen et al. (1999) from the test and rephrased the paragraph to:

"Even though we compare atmospheric measurements of NH_3 concentration from two different measurement systems installed at two different heights separated by 3.2 m (c_{RNH3} in 33 m and c_{dNH3} in 29.8 m), we expect that the measured concentrations are comparable due to the relatively high standard deviation on the concentrations measured by the REA system."

Referee comment #13: *Page 8: Line 20: "2-5C during the rest"*

Response: Has been corrected.

Referee comment #14: *Page 9: Liens 5-9: This is a critical part of the manuscript: The authors should show the trend of the canopy aerodynamic resistance Rac and Rb (as modelled with measured LAI) during the experimental period.*

Response: We calculated both the canopy aerodynamic resistance terms *Ra* and *Rb*, but in order to see the turbulent transfer without seeing the effect of the wind velocity, we calculated the turbulence intensity and chose to use this as this is normalised by the wind speed. As described in the response to comment#1, we investigated whether the leaf fall induced changes in the canopy turbulence transfer, and found that it did not show any trend related to the leaf fall. In the revised

manuscript, we have included a graph showing the canopy turbulence intensity and discussed it in paragraph "4.1 LAI and NH_3 fluxes in the leaf fall period".

Referee comment #15: *Page 10: Lines 4-5: Explain why the model has "difficulties" to handle low friction velocities, and why also it is sensitive to changes in stability conditions (why not only "stability conditions" rather than "changes in . . .").*

Response: The main reason for Gaussian models having difficulties with changes in stability is the assumption of stationary conditions over the modelling period in such kind of models. Additionally, measurements show that the wind direction changes easily at low wind speeds which contradicts the concept of stationary conditions. We have changed the text to:

"It is known that Gaussian models including OML have problems when meteorological conditions change from stable to unstable and low friction velocities prevail (Olesen et al., 2007). One of the main problems is that the Gaussian formulation assumes stationary conditions during the calculation period, which is known not to be appropriate under certain conditions such as low wind speeds (see full discussion in Olesen et al, 2007)."

Referee comment #16: *Page 10 Lines 14-15: But autumn is also a season when leaf absorption is nil.*

Response: The reviewer is correct and this has been specified in the text.

Referee comment #17: *Page 10: Lines 17-18: The observed emission is a combination of increased emissions and/or decreased deposition. The data shown here does not allow concluding. Please moderate the sentence.*

Response: The sentence has been modified to:
"The atmospheric ammonia flux measurements show an interesting temporal correlation with the canopy's vegetation development (Fig. 6A and B) suggesting that F_{NH3} decreases as LAI decreases, and that NH_3 emissions occur in the leaf fall period."

Referee comment #18: *Page 11: Line 16: 'at crucial' should read 'as crucial' I guess.*

Response: This has been corrected.

Referee comment #19: *Page 11: Line 21: I would suggest changing 'in large part' by 'during most'.*

Response: Suggestion has been followed.

Referee comment #20: *Page 11: Line 22: Is evaporation the right term for NH3? What is the mechanism?*

Response: No, the term evaporation is not the right one to use here. It has been changed to volatilization which is the correct term in this case where the NH_3 gas is considered.

Referee comment #21: *Page 11: Lines 27-30: This should be more discussed and also based on Figures of Rac and Rb.*

Response: We have included a short discussion of the canopy turbulence related to the new Fig. 6A in paragraph "4.1 LAI and NH_3 fluxes in the leaf fall period" (see response to comment#14). A part of following sentence has been deleted and a part included in the conclusion (see response to comment#24):

"Beside the forest's phenology, variations in meteorological and soil conditions influence the atmosphere-forest exchange of NH_3 through complex mechanisms of the physical, biological and chemical exchange mechanisms which are controlling the exchange processes."

Referee comment #22: *Page 12: Line 18: Is there any other acid playing a role in this area?*

Response: Although other acids were not measured, nitric acid (HNO_3) could react together with NH_3 to form the particulate ammonium nitrate (NH_4NO_3) (Gallagher et al. 1997). Otherwise, there are, to our knowledge, no other acids playing a role in this area.

Referee comment #23: *Page 12: Line 31: 'gabs' should read 'gaps'.*

Response: This has been corrected.

Referee comment #24: *Page 13: Line 29-30 and 31-32: The causality between observed emissions and litter fall may be a bit more tempered: This is a combination potential litter emissions, diminished leaf absorption, and canopy aerodynamic resistance.*

Response: The sentence has been rephrased to:

"This points to the need for representing forest leaf fall and associated NH_3 emissions in chemical transport models, when simulating nitrogen-deposition to forests. Besides influence on the atmosphere-forest exchange of NH_3 from the forest's phenology, variations in meteorological and soil conditions, and the canopy turbulence, our observations support the hypothesis that NH_3 emission occur from deciduous forests in relation to leaf fall due to a combination of increased litter emissions, decreased leaf absorption."

Referee comment #25: *Table 1 and 2: explain how the uncertainty was estimated.*

Response: In request of referee#1 we have changed the uncertainty to standard deviation in Table 1 and 2. It makes more sense to keep the tables to statistic terms. In Section 2.4 it is written how the uncertainties were estimated and in Section 2.4.1 "Relaxed eddy accumulation" we have specified the process.

Referee comment #26: *Table 2: Give definition of symbols in the legend: CRNH3 CdNH3 CmNH3 and DL*

Response: Symbol definitions is now included in the legend of Table 2.

Referee comment #27: *Figure 3: what does 'Bub" mean? Please explain.*

Response: “Bub” means “De-bubbler”. “Bub” has been replaced with “De-bubbler” and “Mix” has been replaced by “Mixing coil” in Fig. 3.

Referee comment #28: *Figure 4: indexes in ‘NH3’ and exponents in ‘yr-1’*

Response: Has been changed.

Referee comment #29: *Figure 6 and 7: I would suggest joining these two figures and also adding a figure of Rac and Rb on top to help interpreting the data.*

Response: We have joined these two figures in Fig 6 along with a graph showing the turbulence intensity.

References:

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Olesen, H. R., Berkowicz, R. B., and Løfstrøm, P.: OML: Review of model formulation, National Environmental Research Institute, Denmark. NERI, Technical Report No. 609, 130 pp., 2007.

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