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

K. Hansen et al.
krih@ruc.dk

Authors' response to Anonymous Referee #1

We thank Referee #1 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: *Although the paper is well written and interesting, it could be further improved by paying more attention to the sources of these remarkable high ammonia emissions during the fall. At this moment, the authors suggest a relationship with the decreasing leaf area index (LAI) but they only surmise what the controlling factors behind these emissions could be:*

1. *Larger NH3 emission potential Γ of the senescent leaves, connected with remobilization of nitrogen from senescent leaves during the the retranslocation process (page 11, r 5-8)?*
2. *Ammonia emission from decomposing leaf litter (page 11, r 8-13)?*
3. *Volatilization of ammonia from evaporating water film on leaves and moist soil (page 11, r22)?*

*The flux measurements could be compared with bidirectional models in order to better understand the mechanisms behind these emissions. It is currently not clear what the emission potential of the canopy or the litter layer is. The authors applied the deposition model DAMOS, which is, however, unidirectional (using a surface resistance) and therefore inept to explain the emissions. An effort should be done to include/calculate a canopy, stomatal and ground-layer compensation point (see Massad et al., 2010) in the flux modeling.*

Response: First of all, we are very pleased that the reviewer finds the paper interesting. We compare our results to calculations performed with OML-DEP (Olesen et al. 1992, Olesen et al. 2007, Sommer et al. 2009), which is a state-of-the-art local-scale Gaussian dispersion-deposition model, to assess the magnitude of the contribution of the emission fluxes from the forest. In the DAMOS system (Hertel et al. 2013, Geels et al. 2012) OML-DEP is coupled to the regional scale model DEHM, in order to include both the local component as well as the regional component of the $NH_3$ emissions and concentrations. It is true that both models are unidirectional and based on a resistance type deposition scheme, and we are aware that DAMOS thereby cannot be used to estimate the natural $NH_3$ emissions. However, in combination with the measurements, we believe that the DAMOS calculations may be used in the analysis of the potential ammonia emissions from nature. One example is that before leaf fall the modelled concentration level corresponded fairly well to the measured level while it was underestimated for the time period during and after leaf fall. This suggests that the emissions that have been observed in our field experiments are due to processes that the deposition/emission model within DAMOS does not include, indicating that natural emission processes taking place. We agree that an even better understanding of the emissions can be achieved by including a canopy, stomatal and ground-layer compensation point model in the analysis, and this is also planned as the next step in the analysis of the experimental data. However, currently such models (i.e. Massad et al. 2010), that are highly adapted to the specific site, have, to our knowledge, only been applied for agricultural croplands and intensively managed grasslands. The development of a compensation point model for the forest ecosystem is beyond the scope of the current paper. Furthermore, we disagree with the reviewer that a bi-directional model, as exemplified with one by Massad et al. (2010), is a needed requirement in the specific analysis. By definition, the main purpose of compensations point models is to use the stomatal flux of gasses to take into account the overall flux of ammonia through the leaves. However, according to Fig. 1 in our manuscript, the LAI was zero during most of the observational period. This means that there is no respiration within the tree leave canopy and thus no stomatal flux due to respiration. A much simpler approach is therefore is to use a simple unidirectional decomposition model that handles the emission from natural ecosystems such as forests. Such a model can be directly implemented in the existing emission model (Skjøth et al. 2011) that is used in both OML and DEHM (Brandt et al. 2012) within the DAMOS system. The main requirement for such a model is a good estimate of the total nitrogen content in the leaves and a robust methodology to for an accurate description of leaf-fall during autumn, which in current CTM models is still lacking (Sakalli and Simpson 2012). If such a methodology can describe the majority of the ammonia emission flux from forests during autumn, then such a methodology is in general preferred in relation to much more complex models. The reason for this is that modelling of pollution transfer between the biosphere and the atmosphere must be simple enough to ensure reasonable model run times and complex enough to incorporate the key drivers (Sakalli and Simpson 2012). This is supported by recent investigations by Wang et al (2012) that suggest limited ammonia emission from leaves and needles during growth, that the N-content in the leaves were more or less constant during the growing season and that a major fraction of the nitrogen in the leaves was remobilized from the leaves to perennial woody parts at senescence. The amount of remobilization does however seem to vary considerably between species. Our study in combination with the studies by Wang et al (2011, 2012) and the comments by Sakalli and Simpson (2012) therefore suggest that the simpler uni-directional methodology for quantifying ammonia emissions from forests in CTM models should be preferred. Here the main requirement is to obtain the species dependent remobilization fractions of nitrogen and connect this information with a tree species inventory as given by Köble and Seufert (2001) or Skjøth et al (2008). This question is also related to comment #4 from this reviewer.

Additionally, the manuscript has been proof-read by a fellow scientist and we have therefore made additional changes with respect to specific formulations in the manuscript and in order to highlight the relevance of ammonia emission in CTM modelling as well as state-of-the art, we have therefore also expanded the conclusions (page 15649, line 8-10) from ...."

"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. These observations…"

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. Currently a dynamical approach for simulating ammonia emissions are underway in the CMAQ, DEHM, EMEP, CHIMERE, and LOTUS-EUROS models (Aas et al, 2012, Skjøth et al, 2011, Cooter et al, 2012, Hamaoui-Laguel et al 2012). Here calculations of the ammonia emissions as function of ambient conditions are expected to improve calculations and understanding considerably (e.g. Hendriks et al, 2013). But to our knowledge, these methods do not include ammonia emissions from litter fall from forest. Our observations..."

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: *Introduction: page 3,r18-21: please add cuticular desorptions (and reference) for other possible ammonia emission sources*

Response: Cuticular desorption with the reference to Pryor et al. 2001 has been added.

Referee comment #3: *Leaf area index (page 4, r 13-24, page 9 r 3-5, page 11, r 1-2, page13, r7-8): There is some confusion about the determination of the LAI and the PAI. The plant area index (PAI) consists of a leaf area index (LAI) and a stem area index (SAI). From Figure 1 can be seen that before leaf fall the PAI coincides with the LAI, implying that the SAI equals zero. How do the authors explain this? A good estimate of the SAI in deciduous forests can be obtained from the LAI measurements (licor-2000) during the winter period. From Figure 1 can be observed that the winter PAI (SAI) approaches 1? It therefore never drops to zero as written on page 11, r 17. The authors also mention the "green LAI", which I didn't encounter in scientific literature until now. It was said to be derived from observations of leaf defoliation and leaf fall (page 4, r20-21). Was the specific leaf area (SLA) of the beech leaves known to infer the LAI from the leaf fall?*

Response: We understand the point the reviewer raises about PAI coincides with LAI before leaf fall in Figure 1 and we agree with the reviewer's point that there will at any time exist a SAI. However, according to the algorithm applied in the LAI 2000 calculation, the influence of the stems decreases as the LAI increases. We have therefore made the assumption, that when the canopy is fully developed with green leaves (PAI higher than 4), the stems do not significantly affect the LAI that is included in the description of the $NH_3$ exchange between the forest and the atmosphere. Therefore, the PAI and LAI equal each other when LAI is higher than 4. However, the reviewer is perfectly correct that PAI should never drop to zero as written on page 11, r17. This is a mistake and has been corrected in the reviewed manuscript, and we acknowledge that the reviewer has pointed out this mistake. Finally, the end of the defoliation period (leaf fall) was determined from daily digital photos of the canopy using a camera mounted on top of the tower. From these observations, the LAI 2000 measurements were interpolated between the date where LAI dropped below 4 to the date of leaf fall using linear regression. This has been specified in the revised manuscript.

Referee comment #4: *Decomposition of leaves in the fall:*
*The authors suggest that enhanced decomposition of plant material could explain the emissions on 2-4 and 12-14 November ( r23-24). During these events weather conditions (precipitation followed by dry periods with relatively high air temperatures) were found to be suitable for decomposition of the beech litter. It would be interesting if the authors would add some information about the characteristics of the leaf litter, e.g. C/N ratio or pH of the mineral soil. Is the C/N ratio of the beech litter low enough to trigger substantial decomposition in the fall, even at temperatures below 20_C? Is there a well-developed forest floor (?Mg ha-1) or is the forest floor/organic layer lacking at the measuring site? Are we dealing with an acidic or calcareous mineral soil?*

Response: We agree with the reviewer. Also bearing in mind the reply to comment#1 as well as the hypothesis, we believe that this is a key issue to explore further and if possible contribute towards a sufficient simple decomposition model to be used in CTM models. We have therefore added following paragraph to Section 2.1 Field site and referred to it in the discussion p11 r24-25:

"The soil is brown and consists of Alfisols and Mollisols. Dead plant material consisting mainly of leaves and twigs from the beech trees constitutes the top 0-3 cm. Below is a 10–40 cm deep organic layer. In the upper organic soil layers, the C/N ratio is about 20 and the pH is low (4-5), due to a relatively richness in lime (25–50%) (Østergård 2000)."

These soil conditions presented above describes a good degraded soil. Despite the low pH, the conditions for decomposition where $NH_3$ gasses can be produced and volatilized due to the microbiological breakdown of organic material (Brady and Weil 2007) are relatively good. However, we are aware that the observed NH3 emissions measured above the forest are measured under relatively low temperatures (below 10 degrees Celsius) which means that the decomposition processes are slowing down.

Referee comment #5: *Desorption of ammonia from leaves:*

*Page 10, r 19-20: authors also mentioned desorption of ammonia from senescing leaves as possible ammonia source. Is it conceivable that that leaves in the senescent phase become more hydrophobic and water films on leaves evaporate faster?*

Response: Green leaves are considered hydrophobic due to their water vapour saturation and their cuticle layer. During the senescing process we expect these properties to be reduced as a part of the decomposing processes. Therefore, we would not expect senescent leaves to be more hydrophobic, but oppositely be less hydrophobic.

Referee comment #6: *2.4. Ammonia flux measurements:*
*Calculation of ammonia flux measurement uncertainty: It is not clear for the reader how the measurement uncertainty of the ammonia flux has been calculated (as shown in Figure 6). This should be clarified in the methodology.*
*Does the leaf fall in the beech forest (and the accompanying change in the turbulence characteristics) resulted in thorough changes in the calculation of the b-coefficient throughout the measurement period?*

Response: The text in paragraph "2.4. Ammonia flux measurements" concerning the calculation of the measurement uncertainty has been changed to:

"The measurement uncertainty was determined from the relative uncertainty from three calibration measurements of 25 ppb concentration liquids performed during the measurement period. The detection limit (0.03 µg $NH_3$-N $m^{-3}$) was found from calibration measurements of 0 ppb concentration liquids and only a few measurements on 25 October was found lower than the actual limit of detection and then sorted out."

The b-coefficient is not determined from the turbulence itself but from the probability distribution of the turbulence (w). We did not observe any significant changes in the b-coefficient calculations throughout the measurement period caused by the leaf fall.

Referee comment #7: *Table2: change uncertainty by standard deviation _?*

Response: The uncertainty has been changed by standard deviation in Table 2. The same is done for Table 1 in order to be consistent throughout the paper.

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