We thank Leiming Zhang and the two anonymous referees for their thorough reading of the manuscript and their constructive and thoughtful comments which have helped to improve the manuscript further.
All three reviewers judge the manuscript to be interesting and original and worth of publication in ACP. Here we address the individual comments in turn and describe how we have implemented them in the revised manuscript. The original text of the referees is printed in italics.

**Referee #1**

*1. This paper presents a very substantial contribution to the field of biosphere/atmosphere pollutant gas (ammonia) exchange modeling and as such is well worthy of publication in-, and within the scope of, Atmospheric Chemistry and Physics. This significant advance in flux modeling has been eagerly anticipated and awaited in the atmospheric NH3 scientific community for the last decade. The paper is clearly laid out and well written, although excess detail can render some of the figures rather difficult to read and interpret. The literature review is extensive and comprehensive, and the proposed parameterisation follows logically from the review and discussion of the various items in succession.*
*Much effort has gone into synthesizing existing knowledge and into deriving empirical relationships from measured data, even if the authors appear sometimes to have taken shortcuts to alleviate the lack of data in some areas, or to have been selective in the data shown on figures or used in deriving functional relationships in other areas, without necessarily explaining why given data were discarded. This is no doubt a result of the wide range of measurements, methods, techniques, ecosystems, soils, model parameters, etc... covered in the paper and in the large body of literature cited, and simplifications are necessary at this stage to bring the bulk of the knowledge on the topic into one coherent modeling framework, which can be tested, refined and expanded in the future.*

More explanation was added to some of the figures and Figure 5 was simplified by removing numbers relative to references.

*2. The title refers to the exchange between vegetation and the atmosphere, and the emphasis is certainly placed on exchange parameters in plants (stomatal compensation point, leaf surface resistance, bulk ammonium content), while much less space is devoted to the exchange with the underlying ground surface, soil and leaf litter, even though many publications have shown that the magnitude of soil exchange can be similar to, or exceed, that of vegetation. Again, this reflects the current state of knowledge and the lower number of publications regarding soil/litter processes with respect to atmospheric NH3, which by comparison with stomatal and leaf surface exchange, are little known and poorly quantified. Yet there is little doubt that over fertilised systems, the net annual NH3 exchange is largely dominated by soil emissions that occur following the application of fertilisers and manures, and it is clear that this model offers a rather coarse treatment of these emissions. Having said that, there really aren't any operational alternatives, and the present approach is a first step that should be tested at regional and national scales, rather than at the field scale, where comparisons with flux measurements would likely be less favourable.*

We agree with the referee. The soil and litter emissions are a little "under developed" in this paper. Point also made by referee #3. This is due to the lack of data. Nevertheless a more detailed discussion on limits and validation of Equation 20 was added (see reply to general comments #4 referee #3) as well as a table showing $\Gamma_g$ measurements.

*3. The most innovative aspect of the paper is undoubtedly the formalisation of the functional linkage between the (background) stomatal compensation point and the pollution climate as represented by atmospheric N deposition. There is a need to relate the emission/deposition potential of unfertilised, semi-natural ecosystems to the ambient N pollution, and this must be done dynamically to allow ecosystems to respond to changes in emissions and deposition patterns; the model is therefore a useful tool to explore scenarios. However, there is much unexplained variability in the gamma_s datasets and the estimation of N inputs on the basis of bulk NH4+ content in Tables 2, 3 and Fig. 5 for laboratory studies was rather speculative and not necessarily entirely convincing. It may have been safer to limit the analysis to cases where reliable estimates of both gamma_s and N deposition were available (the comment also applies to Fig. 4).*

We agree with reviewer #1. However, if this was to be done the dataset would be restrained dramatically and the criteria for choosing those data points (concerning $\Gamma_s$ measurements) would be a little subjective. We tried however (figure not shown) to exclude data points where N inputs were calculated from bulk $NH_4^+$ measurements (i.e. laboratory studies) and the adjusted curve between $\Gamma_s$ and N input was not significantly different from the one proposed (including all data points) for managed ecosystems. Concerning unmanaged ecosystems some of the data points were already excluded from the data analysis. Those were the data where N input exceeded 50 kg $ha^{-1}$ $yr^{-1}$ based on the argument that unmanaged ecosystems were rarely subjected to such important loads of N input. This data selection was also criticised by reviewer #3. To remove the ambiguity we will exclude all data points concerning unmanaged ecosystems that were done in the laboratory and where total N input was estimated based on bulk tissue $NH_4^+$ measurements. The resulting fitted curve between N input and $\Gamma_s$ for unmanaged ecosystems is therefore the following:

$$\Gamma_s = 246 + (0.0041) \times (N_{dep})^{3.56}$$

*Specific comments*

*1. p10342, l19: ‘too complicated to be integrated in large scale models’: the issue here is not complexity but computing time. Please rephrase.*

Sentence was changed to “they require excessive computing time to be integrated in large scale models”

*2. p10346, l18-21: by ‘total resistance to NH3 exchange within the cuvette’, do the authors mean the artifact in the quantification of the plant/atmosphere exchange due to NH3 being adsorbed/desorbed by the cuvette walls? This should be made more explicit as it represents a potentially important source of error in Xc, Rs, and Rw*

By total resistance to $NH_3$ exchange within the cuvette we mean the sum of $R_a$, $R_b$ and $R_s$ or $R_w$ for the mini-canopy system inside the chamber. In those cases calculating $R_a$ and $R_b$ from cuvette measurement is not as straight forward as for measurements done in the field and could be an important source of error. This was added to the text.

*3. p10347, l12: suggest change to ’...can be attributed to the litter or to the soil, or both’*

Sentence was changed as suggested.

*4. p10348, l15: '...forest ecosystems tend to have low Rw, whereas highest Rw are reported for agricultural grassland ecosystems' : this statement seems to imply that Rw(forest) is lower than Rw(grass), but it is not supported by the data in Table 1, where the Rw(min) values for forests are 3.2, 24, 71, 26,0.1, which gives an average of 25 s/m (geo. mean =7), while values for agric. grasslands are 30, 5, 20, 10, with an average of 16 (geo. mean= 13). The range is wider for forests than for grasslands, but the means are not significantly different at 95%. Further, Figure 7 actually shows that the fitted Rw curve is well above that for grass, regardless of the NH3 to acids ratio. Please rephrase.*

We agree with the referee and the sentence was removed.

*5. p10348, l14-21: were the Rw data presented here normalised to a common reference height?*

$R_w$ presented here are for a height z0 which is variable with vegetation height (as reported in the different studies).

*RH is always higher at the leaf/canopy level than in the air in the surface layer above vegetation, and this certainly accounts for some of the differences in published parametrisations*

We agree that RH is always higher at the leaf level which might explain some of the variability. The ammonia concentration is also different at the canopy level which might explain the variability in the response of $R_w$ to the acid/$NH_3$ ratio. Accounting for leaf level concentrations was not possible in all the cases of this dataset due to the lack of information. This was added to the discussion on $R_w$ paragraph 2.2.

*6. p10349, l6-12: '...evaluate if an obvious trend can be observed': Can one? Is there one? Please comment on Fig. 2. What does it tell us? (it is rather confusing and difficult to read)*

The discussion is more detailed in the other sections of the paper specifically in relation to Figure 7. However additional comments were added with reference to Figure 2.

*7. Figure 3, and p10350, l4-8: I am not sure of the value of this figure, as it is only mentioned briefly in the text and not commented upon. Is it meaningful for example that in the case of semi-natural and grassland, most data obtained by extraction are much lower than data obtained by gas measurements? Does it point to a methodological bias, or is it an indication that that plants used for bioassay were grown on less N that those plants in the gas exchange experiments (either cuvette or micromet.) ? Why is there by contrast a similar distribution of gas and extraction data in crops?*

The authors of the datasets for semi-natural ecosystems (Hill et al. 2001 for example) detect a systemic difference between the two measurement techniques (cuvette vs. extraction) other authors find the opposite (Massad et al. 2008) whereas more recent measurements do not detect a systemic difference between the two measurement techniques (Wang et al. 2010, NEU-Solothurn conference). A comparison for the data set as whole on the other hand might be trickier since different experimental conditions, fertilisation rates and measurement techniques were used rendering the comparision very difficult. The question relative to the difference between grasslands and semi-natural ecosystems on one side and croplands might be indicative that in croplands usually high $N_{input}$ are more common therefore resulting in high $\Gamma_s$ measurements. Comments relative to this were added to the text.

*8. p10350, l27: 'cutting seems to affect gamma_s...': I believe the authors mean here that cutting affects the canopy compensation point (Xc), as the non-recapture (Rw term) of NH3 emitted by litter affects Xc, not gamma_s, and the higher temperature (item ii) does not affect gamma_s itself (since it is temperature independent by nature) but the stomatal compensation point Xs. By contrast, the third item (iii) on plant metabolism is a valid and legitimate argument in favour of cutting affecting gamma_s.*

We agree and $\chi_s$ was replaced by $\chi_c$.

*9. Tables 2 and 3, and throughout the MS: I have a strong objection to using the terminology 'N deposition' for experiments in pots or greenhouses, where plants are grown on either soil or hydroponics and where the N status is controlled, especially for fertilised plants (Table 3). I find it odd for example in the 5th line of Table 2, with beech being grown in a greenhouse on a solution of NH4+ and NO3-, that one should talk of N deposition data (provided by EMEP) while plants grow in the controlled environment of the GH and do not experience atmospheric deposition. (Even if they did so prior to being placed in the GH, is the info still relevant when a nutrient solution is added for 3 months?). I do understand the value and need of compiling data from different sources and measurement methods, and the need to standardise data in order to derive functional relationships, but I think the term 'deposition' should be reserved for outdoor situations. For the present exercise the generic term 'N input' should be used, for semi-natural (Table 2) as well as managed (Table 3) ecosystems, and for outdoor observations the measured or modeled atmospheric deposition can be mentioned separately.*

We agree for indoor experiments the term N deposition should be replaced by N input. We added N deposition values for the Gessler et al. (2000) study (line 5 in table 3) since very similar compensation points were obtained during this experiment for beech trees both in situ (forest) and in the lab.

*10. Figure 4, and section 3.2.1: I agree that for grasslands and crops taken as a whole, the gamma_s data seem to grow exponentially with bulk NH4+. However, there also seems to be two separate populations in Fig.4a, with the grassland gamma_s data being generally higher than cropland data for a given bulk NH4+ level above 20ug/g. Is there anything in the physiology of grasslands compared with crops that might explain this? Could there be a case for splitting the datasets and deriving a (possibly linear) fit for each vegetation type? This would also help reconcile the data by Mattsson et al 2009 with the rest of the grassland data.*

We agree that some of the grassland data points are separate from crop data points this is particular to 3 data points which are from the Mattsson et al. (2009) study. There are however 3 grassland data points that are within the crop data points variability. We don't think it is justifiable to have two parameterisation based on this.

*11. Section 3.2.2: this section does not deal with seasonal variations in gamma_s but annual or longer-term values. Please change section header accordingly. Also, please replace 'atmospheric deposition' and 'Ndep' by 'Ninput' in the context of laboratory studies throughout the section (eg p10352 l23, p10353 l5 and l8, etc), as recommended above, and since Figure 5 itself uses the term Ninput, not Ndep.*

The title to Section 3.2.2 was changed to "dependence of long term $\Gamma_s$ values on N inputs". N deposition was changed to N input wherever necessary throughout the manuscript.

*12. p10352, l18-19, 'gamma_s increases almost exponentially with N input': this statement should be rephrased since the legend of figure 5 mentions that the best fit was a power law (thus not exponential).*

Sentence was changed to: "We notice that $\Gamma_s$ increases considerably with N input (power function)."

*13. As it stands, Figure 5a/c does not show much of a relation- ship unless the green symbols (N inputs derived from lab experiments and bulk tissue NH4+) are removed. The authors argue that for these data, most of the gamma_s values fall below the range of field data (p10353, l6-7) and they should be excluded as the laboratory was unrepresentative of field conditions. This is probably true, but why is not there a relationship between N input and gamma_s for the laboratory dataset by itself, as gamma_s seems rather independent of the added N? Could this point to a very large uncertainty in estimating the N input on the basis of bulk NH4+ content and vice-versa ?*

A relationship between $\Gamma_s$ and N input for laboratory experiments dealing with semi-natural vegetation could be estimated. This relationship would not be very useful in the scope of this paper i.e. proposing a parameterisation to be integrated in CTMs as this case in seldom encountered. We however chose to exclude those data points for the curve fitting here as recommended in a previous comment.

*14. These data based largely on the work in the British Isles by Pitcairn et al (eq. 6) may not be universally applicable. Perhaps, therefore, the relationship between gamma_s and N dep is not as robust as the relationship between gamma_s and bulk NH4+ (p10352, l7-8) , if the N input is rather uncertain.*

We agree that the relationship between $[NH_4^+]_{bulk}$ and $\Gamma_s$ is more robust and this is why this relationship should be privileged where this type of information is available (crop models for example). However in the view of integrating the bi-directional exchange model in CTMs a link to accessible data should be made and hence the parameterisation as a function of $N_{dep}$.

*15. p 10352, l23: 'Ndep values [from the EMEP model] on a grid basis': this is an additional source of uncertainty as N deposition to forest within a grid square will be very different to deposition to short semi-natural vegetation or to managed ecosystems.*

We agree that this is a major uncertainty in the $N_{dep}$ values. We could not however have access to $N_{dep}$ values within each grid attributed on an ecosystem basis. This was added to the text.

*16. p10353, l10 onwards, Table 3 and fig. 5b/d: for managed systems, it seems that all gamma_s values above around 1000 were removed from the dataset to draw Fig 5b/d.*
*This represents 10-12 data points from the upper range of gamma_s values for crops from Table 3 (on page 10387). Could the authors explain why and provide the rationale for the data selection? A number of these data points would undoubtedly weaken the relationship by providing more scatter in the top left-hand corner of Fig. 5b/d, as total N inputs for these high gamma_s data ranged from 0 to 220, with many rather low input values.*

Figures 5 b/d shows $\Gamma_s$ vs. total N input, where total N input is the sum of Equivalent N fertiliser (Table 3 column 10) as calculated from the N status of the experiment and N deposition (Table 3 column 7) as calculated from $[NH_4^+]_{bulk}$ or measured for field experiments. For some experimental data, we could not calculate the equivalent N fertiliser from the N status (when this was provided in mol N/m$^3$ for example), for others we could not calculate the 'equivalent' N deposition (laboratory experiments where $[NH_4^+]_{bulk}$ was not measured). If either of those two data were not reported (columns 3 and 7) then the total N input could not be calculated and thus the datapoints were not shown on Figures 5b/d. This is unfortunately the case for the 10-12 data points from the upper range of $\Gamma_s$. This explanation was added in the text.

*17. Table 3: I counted 36 data points for oilseed rape and 20 data point for barley, out of 69 references in this table for crops. Is not there a potential bias in the parameterisations in favour of these two species compared with other crops eg wheat (only 4 points) or maize (2 points)? I acknowledge that the authors of this MS cannot be held responsible for the distribution of NH3 studies across the scientific world, but could the data be (were they?) weighted to reduce the risk of a bias?*

Data were not weighed to reduce the risk of the bias. Based on your suggestion, we tried to derive a parameterisation by weighing the data according to species distribution. The resulting parameterisation is not significantly different from the original one proposed. Given that some uncertainty lies in the choice of the weighing variables and that some crops are not represented in the data set but we propose to apply the parameterisation to them, we chose to keep the original parameterisation.

*18. p10354, l6: Suggest replace '...linked to annual N input for periods greater than 2-3 weeks after a fertilisation event' by '...linked to annual N input for periods outside of the first 2-3 weeks following a fertilisation event' , if this is what is meant by the sentence?*

Yes this is what was meant. The sentence was replaced accordingly.

*19. p10356, l14-16: the impact of grazing also depends on whether animals are fed solely on grass within the field (in which case there is a net removal of N from the soil/plant system which is converted to animal tissue, meat, wool or milk), or whether the animals are also fed concentrates and other forage on site or in the stable eg during milking twice daily, in which case there may be additional N inputs to the soil/plant system by deposited urine and faeces.*

We agree that this affects the total N budget of the field. In the case of ammonia emissions as stated lower in this review, the biggest emissions in the case of grazing originate from the fact that animals transform the plants (not an important source of ammonia) into urine and dung (much more important source). This was added to the text.

*20. Section 3.2.3: the authors attempt to calculate the N fertiliser equivalent of the NH4+ concentration in nutrient solutions of hydroponic systems. However, in the case of fertiliser and manure applications in the field, significant NH3 losses by volatilisation occur during the first few days, so that the effective or actual fertiliser/manure N input to soil (ie the N that remains available for plant growth) may be of the order of 20-30% less than the N applied. The NH4+ concentration of hydroponics should perhaps be compared with the effective N input by applying an NH3 loss factor (though only for datasets obtained in field conditions).*

We agree with the referee that during fertiliser application there is a 20-30% loss but the same argument can also be applied to a hydroponic solution since there could be $NH_3$ volatilisation from the solution itself.

*21. p 10361, l2: suggest add '...and to set Rbg to zero.' at the end of the sentence.*

The phrase was added as suggested.

*22. Table 5: why should the canopy height and z0 be constant throughout the year for crops (outside the mediterranean and tropical zones)? Clearly there is an annual cycle in canopy height and roughness length as there is for LAI, with either bare soil or eg short (5 cm) winter wheat/barley seedlings or cover crops during winter, and subsequent growth in spring to reach the annual maximum canopy height (1-2m) in summer.*

We agree with the reviewer and changes were made to the table.

*23. p10361, l19-23: '...N input via fertilisation or grazing : the two processes should not be mentioned and treated in the same vein. Fertilisation does add N to the plant/soil system, but grazing (the removal of plant material by herbivores) is a net loss as some of the N ingested is converted to animal protein. Much of the ingested N is indeed lost in rumen fermentation (NH3 volatilisation) and in urine and dung, which return to soil and is available for plant growth. Although grazing is not per se a net annual N input to the system, it may nonetheless locally increase soil available N and thus raise plant N content and gamma_s.*

We agree that this should be added to the text and argued that grazing enhances $NH_3$ volatilization by converting part of the mobilized N in the grass into a potentially high volatilisation source (urine and dung).

*24. p10363, l20-23: I am not of the opinion that 'in the case of significant rainfall, most of the N in the fertiliser is lost in leaching'; the fraction leached depends on the form of the added fertiliser and on soil type and cation exchange capacity. For ammonium nitrate some of the nitrate may be lost but much of the ammonium will remain in the root zone; for slurry, where much of the N is NH4+ there is much less leaching. Incidentally, even if all of the applied N was lost in leaching following rainfall, the parameter T (efolding time) should not be set to infinity (this would mean that gamma_g would never decrease) but to a very small value (eg a few hours). Please change relevant text accordingly and also in Table 7.*

Concerning rain effect and N leaching, unfortunately, we do not have enough data with compensation point type measurements to be able to assess the effect of rainfall. We agree that the amount of N leaching depends on the fertilizer type and too much uncertainty lies behind removing the N applied after a rainfall. We therefore remove this condition and suggest applying equation 20 with care in the absence of additional data.

*25. Equation 20: what does hm mean with a value of 10000 m?*

$h_m$ is a unit conversion parameter to change from hectare to meter. This was added to the text.

*26. p10364, l5: add 'at field capacity' after 'water content'*

Phrase was added as suggested

***Technical corrections***

*Abstract, p10336, l10 : change 'solubility' to 'dissociation'*
*Abstract, p10336, l16 : delete 'set'*
*p 10344, l10: suggest replace 'developed world' with 'Europe and N. America'*
*p10346, l7: replace 'implied' with 'thought' or 'believed'*
*p10347, l13: '...ground layer emissioN potentialS'*
*p10350, l21: change 'Measurement results...' to 'Measurements...'*
*p10352, l22: change 'are' to 'is' p10354, l24: change 'responds' to 'respond'*
*p10359, l18: add a comma after 'permitted' p10360, l16: Suggest add 'ground' to '...for the ground boundary layer resistance...'*
*p10363, l3: 'shortening' -> 'shortage' or'scarcity'*
*p10363, l18 'fertilisers' -> 'fertiliser'*
*p10363, l22: 'following' -> 'follow'*
*p10364, l1: delete 'fertiliser' after 'gamma_g(max)'*
*p10365, l16: 'wetability' does not appear to be an English word (neither in Cambridge nor Oxford dictionaries)*
*p10366, l2: change 'a' to 'the'; delete 'type' before 'model'*
*p10368, l23: the what? funded by the EC NEUIP*

All technical corrections were accounted for.

**Referee #2 (L; Zhang)**

*The main purpose of the present study is to propose a generalized two-layer ammonia bi-directional exchange model for applications in chemical transport models. To meet this goal, a detailed review of compensation points and parameterisations for the cuticle resistance (and other resistance components) were first conducted. Formulas and input parameters for the various components of the two-layer model were then proposed.*
*The paper is generally well written and easy to follow. The materials presented here are useful towards improving the representation of bi-directional exchange of atmospheric ammonia in chemical transport models, but the authors do not address the practical problems concerning the incorporation in such models as the availability of data. I also have a few scientific concerns that worth to be considered.*

We agree with the reviewer that practical issues were not discussed in this manuscript. We feel that this falls outside the scope of the current paper since the aim was to review existing data and propose a generalised parameterisation that can "theoretically" be incorporated in a CTM. Although the parameterisation was conceived taking into account "practical issues" such as the availability of certain data (fertilisation dates, doses etc.) they were also mentioned in the limitations part of the discussion on the parameterisation. The next step would be incorporating this parameterisation in a CTM and thus discussing availability of data on a more detailed and model/region specific basis.

*Major concerns:*

*1. Currently only dry deposition of atmospheric ammonia is considered in the majority of chemical transport models. In order to change the deposition process into a bidirectional*

*process, a two-layer model is proposed in this study. The key addition of the two-layer model compared to traditional dry deposition models is to allow bi-directional exchange through leaf stomata and soil. In many cases, the emission from the soil could be higher than the emission from the stomata. The present study downplays the role of soil emission (and uptake) since nothing on Xg is mentioned in the abstract. The authors have chosen to ignore Xg, in some cases, based on the assumption that the emission from the soil can be either fully or partially captured by the above canopies.*
*Note that the recapturing process is actually built into the equations for Xc, X(z0), and Ft. Thus, theoretically, Xg needs to be included in the model (if soil emission is not negligible) even if soil emissions are recaptured at the above layers.*

The lack of data is the primary reason why $\Gamma_g$ is ignored in managed ecosystems outside fertilisation events and in unmanaged ecosystems. Another reason for this is that most of the compensation point measurements for this type of ecosystems are measurements which do not differentiate clearly between $\Gamma_s$ and $\Gamma_g$ and therefore reflect in some of the cases a measurement of $\Gamma_c$. We therefore propose setting $R_g$ to infinity in some cases to transform the two-layer model into a big leaf model thus limiting the uncertainty both related to the quality of the measurements and the lack of data. Therefore the deposition and or emission to/from the soil is not null but just integrated in the total flux to the surface. This might not be very clear in the manuscript text as it was also pointed out by referee #3. We added those arguments was to the text.

*2. In-canopy resistance to the ground (Rg): In a few places, it is stated that Rg can be set to infinity in order to limit soil emission (if soil emission is not important). However, if soil emission is not important, then deposition to the soil can become important.*
*Setting Rg to infinity will not only be assuming that there is no emission but it will also be assuming that there is no deposition to the soil surface. This assumption is certainly not acceptable considering that ammonia can deposit to any surface quite rapidly.*

Please refer to comment 1 above.

*3. The purpose of an emission/deposition model that is to be implemented in chemical transport models is to produce reasonable flux exchanges above the canopies. Existing dry deposition models have tried to quantify deposition through different paths. The parameterisations for the different resistance components developed in these models have been evaluated with measured total fluxes (e.g., daytime fluxes to stomata, cuticle, and soil surfaces, and night time fluxes to cuticle and soil surfaces). It is quite possible that these parameterisations might underestimate fluxes along one path but these estimated fluxes can then be compensated for by another path. The proposed model picks up parameterisations from different sources for different resistance components. How can we know that the combined resistance parameterisations (the whole model) will perform reasonably, especially over so many different vegetation types? The model as a whole is not evaluated using plant-scale data (despite the fact that the authors have a large data set), nor is it compared with the general overview of the data from the literature.*

Most of the parameterisations of resistance components ($R_a$, $R_b$, $R_s$ and $R_g$) proposed in this paper are taken from Nemitz et al. (2001) in which the parameterised model is compared against crop (oilseed rape) $NH_3$ emissions. Models using similar parameterisation were also successfully applied to grasslands (Flechard et al. 2009) and to forests (Neirynck and

Ceulemans, 2008). This resistance parameterisation is also similar to the resistance scheme used in the EMEP model (excluding cuticule resitance).
Concerning the model as a whole we do not evaluate it against the existing data set since it is this dataset that is used to derive the parameterisation. It is however considered that the model be integrated in the EMEP modelling scheme in the future and the simulated $NH_3$ concentrations be compared to the EMEP measure network in a separate study.

*4. The main goal of the present study is to propose a bi-directional flux model for applications in chemical transport models. A large amount of information that is needed as input for the proposed two-layer model will not be available in chemical transport models at the model grid scale (although it might be available at the plant scale). For example, few chemical transport models have information on fertilization periods, which is key for the Xs and Xg formulations in the proposed model here (Section 4.5). Note that it is more important for chemical transport models to produce long-term average fluxes (e.g., N budget on seasonal and annual scales) and over large areas (e.g., regional scales) than on daily bases. Should the model use more common input information so that the modelling community can benefit from this work?*

This is certainly a major barrier in implementing this parameterisation in CTMs. Even though few transport models include such information, it could be added to the models as input spatialised data. Some datasets are available on a national basis and are derived from census data (e.g. UK, Denmark). Data on a 1 x 1 Km grid base are also available based on a spatial dis-aggregation of estimated application rates at the regional level from the CAPRI regional data base for example for Europe (Leip et al. 2008). The major added value of the proposed model is the mechanistic linkage between the $\Gamma$ and N deposition but also to agricultural practices (which are one of the main drivers of $NH_3$ emissions). This parameterisation allows ecosystems to dynamically respond to changes in emissions and deposition patterns.

*5. Section 2 reviews the modelling approaches (and parameterisations for the different resistance components) and then Section 3 reviews the resistance components again. I feel that these two sections could be better organized.*

Sections 2 and 3 were re-organized as suggested.

*6. A large portion of this paper focuses on cuticle resistance parameterization (Rw). The factors included in the proposed parameterization are certainly very important.*
*One important factor that is not mentioned here is friction velocity (or turbulence intensity) which can sometimes play a dominate role on Rw (as can be see from a multi-layer model of Baldocchi, 1988 and a big-leaf model of Zhang et al., 2003).*

The reason why we do not propose a parameterisation of $R_w$ as a function of U* is that it impacts $R_w$ indirectly via its presence in the parameterisation of $R_b$ and thus changing the surface concentration of the concerned gas.

*7. What are the advantages and disadvantages of this model compared to a few other similar practices that have been done recently, e.g., Cooter et al. (2010); Zhang et al. (2010)? Or at the very least, what are some discussions on the differences among these studies?*

The major added value of the proposed model is the mechanistic linkage between the $\Gamma$ and N deposition but also to agricultural practices (which are one of the main drivers of $NH_3$

emissions). This parameterisation allows ecosystems to dynamically respond to changes in emissions and deposition patterns which is not the case in the parameterisation proposed by Zhang et al. (2010). However, as outlined above, a main drawback of this parameterisation approach is the availability of appropriate input data especially concerning agricultural practices both on a spatial and temporal scale. This discussion was added to section 5.

*8. Conclusion: How big of an impact will be expected from the new proposed model on the air quality model output?*

It is difficult to quantify at this stage the impact from including a bi-directional exchange module for ammonia on air quality model outputs. Several studies conducted at smaller scales and evaluating the impact of including or not bidirectional exchange schemes in transport models show that there is a reduction of up to 50% in predicting $NH_3$ deposition 250 meters from a point source between versions of the model that have a compensation point value of 0 and 30 $\mu g\ m^{-3}$. (Loubet et al. 2009 In Atmospheric ammonia: detecting emission changes and environmental impacts. Results of an expert workshop under the convention on long-range transboundary air pollution).

*Minor concerns:*

*1. Is it necessary to discuss Ra and Rb in detail in both Sections 2 and 4? These formulas are not new and the differences between the different formulas are not large.*

The discussion on $R_a$ and $R_b$ was merged and is now in section 4.

*2. In a previous paper of Nemitz et al.(2000b), different modelling approaches (single and two-layer models) have been discussed in detail. Is Figure 1a still needed here since the paper deals with the two-layer model?*

The paper deals with one layer or two layer model depending on the case. For un-managed ecosystems and managed ecosystems outside fertilisation events we recommend setting $R_g$ to infinity thus transforming the two-layer model into a 1 layer model.

*3. From the definition of Rb (also mentioned in this paper), it should be a resistance at the thin layer above the canopy. Would it make more sense if Rb is in the path above Fs, Fw, and Fg? This way, the formula for Xc can be substantially simplified (see Zhang et al., 2010).*

If $R_b$ was above $F_s$, $F_w$ and $F_g$ this would imply that $R_b$ also applies for bare soil which is not the case. We therefore prefer to keep $R_b$ between $F_s$ and $F_w$.

*4. Tables 5 and 7 provide input parameters for different ecosystems. Do you really think that the information required (e.g., the first column in Table 7) is available at grid scale in common air-quality models?*

Information concerning land use and partitioning between managed and unmanaged ecosystems should be available, However fertiliser input data on a grid scale is not available in common air-quality models. Input maps should be prepared and added as input parameters to the model. Some of the data can be collected from national census in certain countries. Please refer to ‘reply major comment #4’ above.

*5. The paper cited Zhang, Wright, Asman (2010) as 'in preparation'. This manuscript was first submitted to JGR in November 2009 and, as requested, a copy of the submitted version was then sent to the authors of the present paper. I do not think it is appropriate to cite it as 'in preparation'.*

Citing was corrected.

**Anonymous referee # 3**

*This manuscript presents a general parameterization of the two-layer bi-directional NH3 air-surface exchange model. This is a well written paper that will make a significant contribution to the literature and, I expect, will find wide use in the ecological and air quality communities. Given the subject matter, potential impact, and generally high quality of the results, ACP is an appropriate outlet for this work. The authors should be commended for taking on the task of synthesizing the available NH3 flux data for the purpose of constructing generally applicable parameterizations for soil and vegetation emission potentials, as well as the cuticular resistance to NH3 deposition. The paper represents a major first step toward this goal. That being said, the weakness of the paper, which the authors acknowledge, is that for some elements there are not enough data yet available to develop robust parameterizations, leading to large uncertainties.*
*In other cases multiple techniques have been used to collect data, which exhibit some systematic differences, further complicating data interpretation and parameterization.*
*Overall the authors have done a responsible job of presenting the weaknesses and uncertainties of the data and in comparing the resulting parameterizations to field data.*
*However, there are some areas, as described below, where more description and detail are needed. Though some additional work is required, I believe the authors can address these comments. Subject to thorough treatment of these comments, I would recommend publication.*

*General comments:*
*1. Construction of generally applicable relationships between $\Gamma s$ and system total N input is a significant advancement for NH3 modeling. The authors have done a good job assembling and interpreting data from the numerous studies represented in Figure 5. In general I think the resulting parameterizations represent a useful first step and I anticipate they will be widely used. That being said, I do have a few concerns about the methods by which the original data were adjusted for comparison and, in general, the comparability of some of the data. First, the metric on which the $\Gamma s$ parameterization is based is total annual N input. For field studies describing semi-natural systems this value is either given as atmospheric deposition measured at the site or can be derived from air quality models. For arable systems, however, I expect that in some cases the published results only include application rates for the specific growing season under investigation, rather than annual fertilizer input for the site. Have the authors accounted for this in summarizing the field measurements in figure 5?*

No this was not accounted for in crops, some agricultural grasslands have annual fertilisation rates others not. The fertilisation rates accounted for in agricultural crops are the applications for the growing season. This is for 2 reasons: (i) it is very difficult to know the yearly N fertilisation for the data collected from the literature; (ii) it can be argued that managed ecosystems are seasonal or annual crops unlike perennials for semi-natural and forest ecosystems and thus are more affected by the seasonal N input rather than the annual N fertilisation. It can also be argued that the N fertilisation on the previous crop was presumably used by that crop and exported out of the field upon harvest (applies more to crops than grasslands).

*2. Also, it looks as though not all of the data from Tables 2 and 3 are included in Figure 5. I may have overlooked something in the text, but the authors should explain the criteria for including data in Figure 5 and discuss how this may affect the resulting parameterizations for $\Gamma s$.*

This is a major mis-understanding in the paper that should be clarified as referee #1 also points to this. All the data points in tables 2 and 3 were included in figure 5. Some of the data points where the fertilisation rate or the N deposition could not be calculated or derived were not reported on the figure (refer to referee #1 specific comment #16).

*3. It is unclear to me whether the parameterizations resulting from Figure 5 (equations 7 and 8) exclude data from laboratory experiments. Firstly, in either case, I am not comfortable with the term Ndep being used to describe fertilizer application rates in laboratory experiments. This term should only be applied to studies carried out in the open atmosphere. Secondly, the laboratory experiments (green symbols) in Figure 5 a and c show very little or no correlation between $\Gamma s$ and total N input. Why is this? I agree with the authors it is unlikely that this results primarily from uncertainty in estimating N input. However, a more detailed discussion of other potential reasons is warranted.*

The original parameterisation only excludes 4 data points which are for Hill et al. (2001) and Mattsson and Schjoerring (2002). Those points correspond to semi-natural vegetation that was moved to the laboratory and to which excess N was applied daily mimicking high N deposition rates. We consider that it corresponds to unrealistic events and that semi-natural vegetation experiencing high N deposition naturally would have time to adjust accordingly (if not in the lab). Based on the suggestions of referee #1 all laboratory based studies concerning semi-natural ecosystems were not included in the derivation of the parameterisation (but are still shown in figure 5) for the reviewed version of the manuscript (refer to referee #1 comment #13). This clarification was added to the manuscript.
We agree about the confusion created by using the term $N_{dep}$ for fertilizer application and we replaced it by N input.

Concerning the laboratory experiments in figure 5(a and c). Those were excluded from the parameterisation (see above) but probable reasons for the lack of correlation could be the following:
- These data points are in the majority semi-natural vegetation that were transposed to the laboratory and watered with high N solution for a short time interval (Hill et al. 2001 and Mattsson and Schjoerring 2002). One can think that in the case of semi-natural vegetation an adaptation time is required for the high N input to be reflected in $\Gamma_s$.
- These data points are for a range of different plant species and as shown by Mattsson et al. (2009), there is an important interspecies variability in $\Gamma_s$.
- On top of the uncertainty in the estimation of the total N input to the experimental setup there is an uncertainty in the measurement of $\Gamma_s$ especially that this was done with different methods (cuvette vs. apoplast extraction).
These arguments were included in more details in the text.

*4. Understandably, the text is weighted toward the development and discussion of the parameterizations for $\Gamma s$. In my opinion, the treatment of $\Gamma g$ would benefit from more detail.*

*A more complete description and summary of available data, similar to what is done for $\Gamma$s, would add context to the description in section 10363. Furthermore, as I discuss below, the rationale for setting Rg = ∞, and therefore ignoring $\Gamma$g, for unmanaged systems and managed systems with overlying canopy requires a more in depth justification.*

We agree that the section concerning $\Gamma_g$ was a little underdeveloped this is mainly due to the lack of data. We however added some detail to the description of data and included a Table (Table 4) summarising all data available. Justification for setting $R_g$ to infinity in some cases was added to the text. Please refer to comment 11 below.

Specific comments:

*1. 10336, line 6: remove "here"*

Word was removed.

*2. 10347, line 2: "has sometimes provided smaller values than the gas exchange measurements". This statement stops well short of describing the seemingly systematic difference between the approaches for measuring or estimating gamma. A stronger statement or further description is needed*

This sentence was replaced by "Systemic differences between apoplast extraction and the gas exchange techniques were reported (Hill et al., 2001, Massad et al. 2009), these are attributed to errors in both methods and should be further investigated."

*3. 10350, line 2: The authors tend to downplay the disagreement between the 3 techniques for semi-natural systems. The authors should more directly acknowledge the disagreement and provide a brief discussion of the possible reasons.*

The comment in this paragraph refers to the compiled data in this study where we did not detect any difference between the techniques. A comment on the systemic difference between the methods was added in a previous section (2.5).

*4. 10350, line 26: "By contrast, cutting seems to. . . ." Only point (iii) is relevant to $\Gamma$s.*

We agree that the three points are more relevant to $\Gamma_c$ than to $\Gamma_s$ and this was replaced accordingly in the text.

*5. 10350, line 17: "Field fertilizer application results in. . . ..". As a statement regarding NH3 emissions this sentence seems out of place. Are the authors referring to the peak in $\Gamma$s?*

This refers to a peak in $\Gamma_s$ but also in $\Gamma_g$ which most of the time results in a peak in $NH_3$ emissions (fluxes). $\Gamma_s$ is particularly relevant to this paragraph and therefore the sentence was corrected accordingly.

*6. 10362, 1ine 3: Would canopy height be provided as a model calculation?*

Canopy height is not a model calculation in the two-layer ammonia exchange module but is an input variable which can either be provided from input tables or from a coupled model in case the ammonia module is incorporated in a more general CTM.

*7. 10362: The rationale for setting Rg = ∞, and therefore ignoring Γg for unmanaged systems and managed systems with overlying canopy requires a more in depth justification. In managed systems, while the overlying canopy may recapture most of the emissions, the emissions themselves may be large, and therefore significant with respect to the net-canopy scale emissions. In unmanaged systems, particularly forests, the emissions will indeed be much smaller but again may be important in terms of in canopy NH3 cycling, and therefore play a role in the net canopy exchange. I do realize there is a lack of data from which to soundly parameterize these components of the model but my feeling is that Γg for unmanaged systems and managed systems with overlying canopy should not be ignored.*

As the reviewer states, the lack of data is the primary reason why $\Gamma_g$ is ignored in managed ecosystems outside fertilisation events and in unmanaged ecosystems. Another reason for this is that most of the compensation point measurements for these types of ecosystems are measurements which do not differentiate between $\Gamma_s$ and $\Gamma_g$ and therefore reflect in some of the cases a measurement of $\Gamma_c$. Therefore proposing a big leaf model for those cases might be a way to limit the uncertainty both related to the quality of the measurements and the lack of data. This argument was added to the text.

*8. 10363, line 3: the phrase "shortening of data" may be unclear to some readers.*

Shortening of data was replaced by "shortage"

*9. 10364, line 20: I do not agree with the statement that most of the N fertilizer is lost to leaching after the first rainfall, particularly NH4+. In fact, in some cases a second large emission pulse is observed following the first rain event after fertilization, as rainfall directly stimulates chemical transformation of the fertilizer and mobilization into the soil thereby stimulating microbial processing. The authors should acknowledge that equation 20 will not capture such dynamics. As mentioned above in my general comment, the section on the temporal dynamics of Γg after fertilization requires more detail and would benefit from the presentation of graphics demonstrating the agreement between the proposed parameterizations for Γg and field data.*

We agree with referee #3 that equation 20 will not capture the complex dynamics in $NH_3$ emissions from the soil layer. A major variable to account for is the type of fertilizer applied (NO3, NH4, organic, …). We have practically no data showing a second peak in $\Gamma_g$ after a rain event. A more developed discussion on $\Gamma_g$ in general and on the limitations and validation of Equation 20 was added (please refer to reviewer #1 comment 24).

*10. Figures and Tables:*
*NH3 units in Table 1 should be ug/m3*
*Caption for Table 2 should be consistent with the text regarding Rw and Rw(corr)*

Caption and unit were corrected accordingly.