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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Review and parameterisation of bi-directional ammonia exchange between vegetation and the atmosphere" *by* R.-S. Massad et al.

Anonymous Referee #1

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Review of ACPD manuscript:

'Review and parameterisation of bi-directional ammonia exchange between vegetation and the atmosphere' by R.-S. Massad, E. Nemitz and M. A. Sutton

General comments

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.

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.

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).

Specific comments

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

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

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

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.

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

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)

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?

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.

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 NH_4^+ and NO_3^-, 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.

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 NH_4^+. 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 NH_4^+ 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 etal 2009 with the rest of the grassland data.

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.

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). 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 NH_4^+) 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 NH_4^+ content and vice-versa ? 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 NH_4^+ (p10352, l7-8) , if the N input is rather uncertain.

p 10352, l23: 'Ndep values [from the EMEP model] on a grid basis': this is an additonal 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.

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.

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 NH_3 studies across the scientific world, but could the data be (were they?) weighted to reduce the risk of a bias?

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?

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.

Section 3.2.3: the authors attempt to calculate the N fertiliser equivalent of the NH_4^+ concentration in nutrient solutions of hydroponic systems. However, in the case of fertiliser and manure applications in the field, significant NH_3 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 NH_4^+ concentration of hydroponics should perhaps be compared with the effective N input by applying an NH_3 loss factor (though only for datasets obtained in field conditions).

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

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.

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 (NH_3 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.

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 NH_4^+ there is much less leaching. Incidentally, even if all of the applied N was lost in leaching following rainfall, the parameter T (e-folding 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.

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

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

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 NEU-

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