

We would like to thank Frank Dentener for a very constructive and thorough review of this paper. This has resulted in substantial improvement in the paper.

However it would be valuable to provide an estimate of the pre-industrial and present day impacts of this temperature signal (in the order of 0.7 C world average, and higher in the major agricultural production areas) on NH₃ emissions.

We have now produced an estimate of this. To produce this estimate we have run a set of additional simulations to test the sensitivity of the NH₃ emissions and other N pathways to a global change in ground temperature of +/- 1° C. A temperature increase of one degree is roughly equivalent to the observed increase in surface temperature over land since the pre-industrial period. Results show a global sensitivity of about 1 Tg (NH₃)/yr per degree C for the given conditions and manure and fertilizer application rates in our sensitivity simulations. The results of these additional simulations will be included in the revised paper and used to interpret the increase in emissions since the pre-industrial.

Thank you for this suggestion.

Although the comparison looks reasonable (note here the static inventories partly make similar assumptions), I am left with the impression that this agreement is dependent on rather arbitrary choice of key-parameters such as soil pH and canopy capture factor. Especially the assumption that on average 60 % of nitrogen is captured by plants is rather unconstrained and critical. As far as I understand CLM4.5 does consider plant functional types that include also nitrogen pools in the various tissue. In my view this represent a missed opportunity to get more solid insights in the role of soil-canopy exchange in determining emissions.

There is no doubt the canopy capture is rather ill constrained. Stuart Riddick (Riddick, 2012) investigated this as part of his PhD thesis (Chapter 4) using seabird nitrogen as the fertilizer source on different substrates (rock, sand, soil and vegetation) inside a chamber to investigate the ammonia capture as it volatilizes to the atmosphere. This experiment found ammonia recapture to be: rock 0%, sand 32%, soil 59% and vegetation 73%. We chose a value of 60% as it was in-line with the findings of Wilson et al. (2004) and is mid-way between the value for soil (when the crops are planted) to when they are fully grown. Bouwman et al (1997) also used canopy capture to estimate emissions with the captured fraction ranging from 0.8 in tropical rain forests to 0.5 in other forests to 0.2 for all other vegetation types including grasslands and shrubs. He omitted canopy capture over arable lands and intensively used grasslands. The PhD thesis of Stuart Riddick suggests capture may also be important in these locations.

Canopy deposition is also important in the deposition of nitrogen from the atmosphere to the surface. In terms of deposition of nitrogen onto the canopy some measurements are as follows: through an experiment using labeled ¹⁵N Dail et al. (2009) measured around 70% of total atmospheric N application was found on trees (plants); Adriaenssens et al. (2012) reported that only 1–5% of the applied dissolved ¹⁵N was taken up by leaves and twigs, a minor fraction in the high ambient N deposition of north Belgium (approximately 30 kg N/ ha-yr deposition); Wortman et al. (2012) found that the canopy retained 20–25%

of the total atmospheric deposited N at the Novaggio forest, Switzerland (experiencing high rates of N deposition: 25–40 kg N/ ha-yr). In a low ambient N deposition area (<5 kg N/ ha-yr) Gaige et al. (2007) demonstrated a 70% retention of added N in the canopy at the Howland Integrated Forest Study site (east-central Maine, USA), but a later study reported that 10–25% of the 15N was retained in or on twig and branch materials whereas only 3–6% was recovered in live foliage and bole wood; at the Niwot Ridge AmeriFlux site (Colorado, USA) in a low ambient N deposition area (3 kg N/ ha-yr), approximately 80% of the growing season total N deposition was retained in canopy foliage and branches. Global modeling simulations generally ignore the canopy deposition of nitrogen and settle for adding all the reactive nitrogen deposition into the soil (TAN) pool (see especially, Zaehle 2013 and Zaehle and Friend, 2010).

NH₃ is both uptaken and deposited on the canopy. The deposition of NH₃ onto the canopy (or even the soil surface) is poorly constrained (e.g., see Erisman and Draaijers, 1995) and often ignored in model simulations. Explicitly including the canopy capture fraction allows us to explicitly differentiate between different biogeochemical pathways. In the future when the model is fully coupled with the atmospheric ammonia cycle a compensation point approach would be desirable, but we feel it is outside the scope of the present study. The suggestion that our global model could provide greater insight into soil-canopy exchange seems a little brave given our model's uncertainties. However, different pathways will be considered when the nitrogen emissions are fully coupled to the simulation nitrogen cycling.

We address the issue of pH in more detail below under specific comments.

Another issue that can be probably relatively easily addressed is the run-off of N and comparison with other estimates. Once NH₃ is emitted in the atmosphere it doesn't mean it is gone- it will probably deposit for a large part not very far from the sources. Also NO_y deposition will contribute to nitrogen inputs into the watersheds. It would be good if the figures and text dealing with this correct for this phenomenon, or clearly mention that run-off only partly includes the full picture.

This is a good suggestion. We will further clarify in the text that the runoff calculated here only accounts for part of the picture, the reactive nitrogen that is directly runoff from manure and fertilizer applications. Further runoff may be expected downstream resulting from nitrogen redeposition. Nevison et al. (2015) examines the runoff from models that only include deposition and those that include both deposition and agricultural representations. In locations with substantial human impact the deposition from agriculture is by far the dominant component as calculated in this model. A number of fertilization studies have also suggested considerable reactive nitrogen retention from natural ecosystems. For example Nadelhoffer et al. (1999) shows a loss of 10% of nitrogen from nitrogen deposition in temperate forests. This suggests that much of the redeposited nitrogen is in fact incorporated into natural ecosystems downstream and not into runoff.

Sensitivity tests.....but of course it should be recognized that some of the parameters may co-vary and can not be seen in isolation.

Good point. We will mention this in the revised test.

You mention that simulation was run in “decoupled mode” with atmosphere, using reanalysis from Quian et al (2006). As you mention, several important processes of exchange between canopy and atmosphere are not explicitly simulated. This is an important consideration, given that the canopy “health” status and biomass accumulation depends critically on fertilizer input. Nitrogen affects also the other processes in crop canopy, such as transpiration rates, which might have the cooling effect over the cropland. Therefore, fully coupled model should be explored in the future to observe and understand better also the feedback mechanisms related with the atmosphere. Are you planning to perform these simulations?

Yes. We are presently carrying out simulations where the CLM NH₃ emissions are coupled to the atmospheric model so that we can explicitly couple the atmospheric nitrogen cycle to the land model using the parameterization described in this paper for agricultural nitrogen emissions. We hope to be able to integrate a NH₃ compensation point approach into the model to fully investigate these interactions. These type of feedbacks will indeed be interesting to explore. We will include a section on next steps in the conclusions.

I suppose a next important step will be to incorporate agricultural management model, trying to capture these processes better. Nevertheless, due to high spatial variability of agricultural management practices related to fertilization, the results of such simulations might have limited value for regional applications when run on coarse resolution. How do you foresee in the future this problem might be tackled? Perhaps a kind of up-scaling method could be applied to bridge the gap between local (management practices) to coarser resolution of earth system models?

Yes, an important further step will be to explicitly include agricultural management practices along with a better distribution of the fertilizer and manure applications over a grid-cell. However, one problem at present is that several physical quantities predicted by the model that are needed by the N-pathways model (see Table A1), particularly soil-related variables, are not predicted by plant functional type (PFT) in the CLM. Instead these variables (including the ground temperature, soil water content, etc.) are predicted by soil column. Each soil column can contain different PFTs so it is not advantageous to associate N application from the inventories to specific PFTs in this scheme except to include different resistances. However, the explicit agricultural model available within the CLM will be of value in more accurately representing the emissions. Truly regional applications would likely need more complex regional models requiring more accurate representation of explicit regional practices and soil characteristics. The global problem is

difficult, of course, as in some regions agricultural practices are well documented while in other regions they are not. To some extent atmospheric chemistry models run into the same problems with heterogeneous emissions and yet these models when evaluated on the regional scale have proven quite valuable. These next steps will be discussed more explicitly in the paper conclusions.

This spatial heterogeneity also makes an evaluation of the simulations difficult. Measurements of NH₃ concentrations also do not have great coverage in space or time. The use of wet deposition data and aerosol speciation may partly alleviate this problem by providing a wider emission footprint but involves other simulation uncertainties. An evaluation of these type of simulations over well-characterized regions in combination with a field study that has a high concentration of NH₃ measurements would be valuable. An ongoing project using a landscape scale model that couples the atmosphere, plants and a farm decision model carried out on French and European landscapes (Duret et al. 2011, Drouet et al. 2012) may allow us to leverage with existing projects.

Despite our criticisms, we nevertheless think that the study is very valuable since it provides a first framework on top of which in a later stage modifications, improvements and extensive testing will be possible. I think the authors did a good job in describing assumptions, but the manuscript could include some more detailed insights on the next steps to be taken. Unfortunately the manuscript doesn't read very smoothly, any improvement of the readability would be welcome.

We will include our next steps in the conclusion section (many of these are discussed above). We will also try to improve the readability of the paper in the next iteration, although specific suggestions of the sections that proved difficult would be helpful.

p. 15949 l. 13,14,20. It would be useful to have the numbers matching the overall N-inputs. Providing soil Nr formation would be useful. Please keep the same units.

We will include the overall Nr-inputs and soil Nr in the abstract.

p. 15949 l. 20 Why do the earlier mentioned emissions in 2000 not match? In case of all nitrogen I would expect $21+12=33$ Tg N (40 Tg NH₃) if only organic nitrogen: 25.5 Tg NH₃.

This was miswritten in the original manuscript. The new text should read “17% of nitrogen applied in 2000 ($21 \text{ Tg (N) yr}^{-1}$)”.

p. 15951 l. 13 provide year of reference since numbers can change rapidly over time.

These are numbers from the Fowler et al. (2013) for present-day. We will provide more precise dates when available, although in many cases with the wide range of uncertainty the precise date may not be of particular importance.

p. 15951 l. 28 in future

Yes, thank you. We will specify this.

p. 15952 l. 14 degree C.

Yes, thank you. Degrees C.

p. 15952 l. 13-15 it should probably be recognized that also other soil microbiological processes may change with climate, as well as agro-management practice (beyond this study).

True. We will mention.

p. 15959 l. 28 l. 182 check the chronology of the publications, hard to imagine that results that were published in 2010 (and not available in 2007) were used in 2007.

We are not sure exactly what this is in reference to? (Potter et al. (2010) gives values valid for 2007).

p. 15957 l. 14 I agree, but a correct mix of the two concepts is a challenge. This could be further elucidated in the discussion.

We agree. In our new discussion of future plans we will elucidate this in some detail.

p. 15957 I recommend to use the wording synthetic fertilizer (or inorganic fertilizer) as manure is also a fertilizer. Throughtout the manuscript.

Thank you. We will use synthetic fertilizer as was suggested in a previous version.

p 15959 l. 17 at least in Europe import of feed is at the core of many nitrogen-related environmental issues. I propose to delete the 'generally' statement, unless the authors are certain that this is not the case elsewhere.

Agreed.

P 15959 l. 25 I would like to see this discussion on simplifying assumptions such as not explicitly using crops, but rather what is in the land-model, earlier in the manuscript, possibly in the introduction. It is not very clear to me whether there is a consistency between the gridded manure database, and the CLM land-use. How much of the manure is on place where CLM thinks there is a forest, desert or similar? It would be good to know that indeed manure is placed where grasslands and crops grow (+fractions).

Yes, we can move this up. As discussed above several physical quantities predicted by

the model that are needed by the N-pathways model (see Table A1), particularly soil-related variables are not predicted by plant functional type (PFT) in CLM. Of course, the quantities used to compute N pathways can then be “contaminated” by forest or bare ground PFTs that may exist on the same soil column. In practice we expect that the impact of this contamination will be small since the major N-application regions (central US, northern India, eastern China) are not PFT-diverse but contain almost exclusively crop and grass PFTs. Subgrid variability will be explored in more detail in our next iteration of this work and will be addressed in the discussion on future improvements. The introduction of an agricultural model within the CLM framework will address part of the concern here. Incorporation of PFT dependent canopy deposition and aerodynamic resistances are among future improvements.

On a gridcell basis, there are a few regions globally where the Potter et al. (2010) dataset includes manure or fertilizer and there is a zero fraction of crop area in the translation of the Hurr et al. (2011) land cover dataset to CLM PFTs. These regions include boreal Asia and North America and western Australia. In these regions the N applied from the Potter et al. (2010) dataset is quite small in magnitude and these regions are covered mainly in grass or shrub PFTs.

p. 15961 l. 16: it is quite well known that net-losses from urea (NH₃ emissions), can be much higher (15-20 %) than for most other nitrogen components (a few %). See Bouwman et al., (1997) Is it correct to assume that the numbers in this manuscript are from this point of view an upper limit? I am surprised by the sensitivity study which contradicts these general view, and I don't understand why.

The discrepancy in these results may be due to the pH changes in the soil that may occur with the application of fertilizer. Bouwman et al. (1997) appears to have based their results on Whitehead and Raistrick (1990), who conducted laboratory experiments of soil columns with different types of soils and different types of fertilizers. As fertilizer is added to the soil sample the pH changes. Whitehead and Raistrick (1990) find a close relation between the resultant pH of the soil and the amount of ammonia volatilized. In most instances urea increased the pH more than the synthetic fertilizers. How emissions respond to actual field conditions and farmer's practices may of course be more complicated. This is another aspect of the parameterization that can be revisited when agricultural practices are further incorporated.

p. 15961 l. 24; corn (grain maize) is an important crop. However it is not representative for instance for winter wheat, another widely used crop. Some discussion on the implications could be given.

We use the planting date criteria for corn to determine the date of fertilizer application since these criteria lie between the earlier planting for temperate cereal crops and the later planting of soy as given by the CLM4.5-CN crop model. The N pathways are sensitive to the fertilizer application date and rate (Table 2) so this should be considered in future development. Fertilizer application in winter has been shown to be important for estimating NH₃ emissions in some regions (e.g. Paulot et al., 2014) with important

implications for air quality issues. We will add discussion of the implications in the new paper version.

p. 15964 l. 12 There are a number of modeling papers, and satellite retrievals that could provide a more insight on the range of concentrations. See recent review in Zhu and Henze Curr Pollution Rep (2015) 1:95–116.

Thank you. We will include this reference in the revised version.

p. 15964 l. 20 Can the authors comment on the physical evidence for such ‘re-capture’ of NH₃ emissions? It is well known that nitrogen in the plants tissues will also tend to maintain an NH₃ partial pressure. If stomata are closed this will probably less so play a role. Is this mainly a tuning factor? And how sensitive is the model to it?

Plant recapture of emitted ammonia is non-negligible. This is often reported to be as high as 75 % (Harper et al., 2000; Nemitz et al., 2000; Walker et al. 2006; Denmead et al., 2008; Bash et al., 2010). However the stomatal pathway is not the only recapture pathway since NH₃ can be deposited to the cuticle especially at nighttime. However it is our understanding that the fate of the overall biological pathway of nitrogen captured by plants is still controversial within the ecology community. So far we have not explicitly included the biogeochemical cycling of nitrogen from recapture within the model. The ultimate consequence of this recapture is likely to increase the reactive nitrogen in the litter pool and the reactive nitrogen in the soil pools (since the plants will not draw the nitrogen from the soil pools to as large an extent).

p. 15971 l. 18 Descriptions of global maps : : .etc

The global maps are described line 19-25 on 159711.

p. 15973 One aspect which should be mentioned here is that in reality the NH₃ emitted, will probably not travel very far and on the time scale of hours (NH₃) or days (NH₄) deposition near the emission regions. On the gridscale considered, it can be safely assumed that most of the NH₃ also enters in run-off. It was not clear how run-off simulations of this model compare to observations were taken into account (see general remark)

This is discussed in detail in association with the general remark above. In the model evaluations of runoff from fertilizer and manure the atmospheric redeposition of ammonia was not accounted for. We will clarify this point.

p. 15973 l. 25 See previous remark.

We will discuss in greater detail the role of managed agricultural practices for manure.

p. 15973 confusing use of the word deposited – I assume the authors talk about atmospheric deposition.

This refers to atmospheric deposition. We will make this explicit.

p. 15974 I do think the hypothesis of some fraction of NH₃ being readily available to plants could be tested using the information available in CLM. It is a missed opportunity.

Plants can use the deposited N as N enters via the stomata (which can be accounted for if we use the bidirectional exchange scheme). Another part of the captured N will probably be washed by the rain/dew etc to the ground and be added to the soil N pool or the litter pool (Flechar et al. 2013 for a review). A number of aspects of this nitrogen cycling are indeed rather unconstrained. While we could test this in future projects the ultimate fate of this nitrogen is likely to remain unconstrained without additional measurements.

p. 15976 l. 5 EDGAR4.2 output contains output separating manure management and manure directly on the field. It seems that the authors have not included manure management (sector B2), which at least to a great extent finds its way in to the fields as well. This would make the overall EDGAR estimates from agriculture larger, but the ratios more consistent. Beusen et al. 2008 estimate does include emissions from housing/storage.

Thanks for this comment. The EDGAR4.2 emissions are in fact split between agricultural soils and manure management. The notes for manure management suggests it includes the following categories: “Country-specific/regional manure management systems: Pasture/range/paddock, Daily spread of manure, Solid storage, Drylot, Liquid/slurry, Lagoon, Pit, Digester, Burned for fuel, Other” (http://edgar.jrc.ec.europa.eu/factsheet_4a-b-d2.php) while agricultural soils include fertilizer and manure applied as fertilizer (http://edgar.jrc.ec.europa.eu/factsheet_4a-b-d2.php). So the numbers given for EDGAR (figure 11, where the units should be Tg N/year) are not correctly apportioned between manure and fertilizer. The numbers given in this figure for EDGAR for fertilizer apparently include both manure and synthetic fertilizer. However, the totals are consistent with the overall EDGAR totals.

The numbers given for manure in the inventory include both manure management and explicit spreading onto fields and pastures. We assume all manure is spread onto fields and do not explicitly include management practices. An extensive discussion of this is given in response to reviewer #2.

We will correct the figure in reference to the EDGAR inventory as it incorrectly attributes manure emissions to synthetic fertilizer. Moreover, we will include increased discussion as to impact of animal housing.

p. 15978 I am somewhat surprised by the low sensitivity to the background concentration. It would be good to include in the figures a visualization of the NH₃(g) the NH₃ concentration in equilibrium with the TAN pool.

We are glad the reviewer noted this apparent inconsistency – this was a typo, the order of

magnitude of the change in concentration was miswritten and has been corrected. The sensitivity simulation was actually run with a background concentration of 1 ug/m³, not 10 ug/m³.

15970, line 5: Does this imply that other factors, such as wind and water availability lead to increased NH3 emissions, even though the temperature is lower?

All other things being equal, a decrease in temperature will lead to a decrease in the NH₃ emissions, but as shown by the sensitivity study there are many factors that can impact the emissions and make it possible to see higher emissions at lower temperatures in some cases. Riddick et al. (2014, 2015) and Riddick's PhD thesis shows the variability in NH₃ emission from a nitrogen source (seabird guano) in a range of locations and climatic conditions. The measurements presented in this paper clearly show that temperature is not the sole driver of NH₃ emission and emission varies considerably due to other factors (winds speed, water availability, substrate and relative humidity). Also see Søgaard et al. (2002) for factors leading to different emissions.

15970, line 12: Can you clarify why in your opinion the performance over grassland is better? In addition, differences in the distribution of manure over grassland and arable land can affect the NH3 emissions, since the emission factors differ between slurry application methods on grassland and arable land. Is arable land foreseen as category of land use in your simulations? Perhaps, some clarification would be needed.

It seems clear we would capture natural emissions from grassland better than emissions from feedlots. Feedlots are more managed while the described parameterization assumes manure is spread on the land. One of the next steps is to add manure management practices. This will include differences in the use of manure on arable land (handled through the agricultural model in the CLM) and the use of manure on grasslands (which are distinguished through their pft).

15973, line 23: Did Bouwman take into account soil nitrogen pools, which could describe the difference between the two studies?

The reference here should be to Bouwman et al. (2013). Nitrogen soil pools are not explicitly considered as the calculations are through a budget methodology. However, changes in the soil nitrogen budget are considered. The error bars on both calculations are likely very high.

p. 15979/l 10 . On page 15965/l 19 the authors declare that soil pH is simply set to a value of pH=7. The sensitivity tests suggest that pH is one of the more determining factors for NH3 emissions (as expected). Can the authors explain why they have not used one of the global soil pH databases around to try to get more robust results?

We have run an additional sensitivity simulation that includes a global soil pH database in place of the constant values we used before. The data are from the ISRIC-WISE database (Batjes, 2005) and have a global average soil pH of 6.55. This decreased the

manure emissions by 23% and the fertilizer emissions by 14%. We will report these values in the table showing sensitivities in the new manuscript

However, it is not immediately clear what the appropriate pH values should be as planted fields are often buffered for pH and manure has its own pH. Moreover, the pH of the TAN pool may depend on the extent that the fertilizer is mixed into the soil. Moreover, the addition of fertilizer may change the soil pH itself (Whitehead and Raistrick, 1990). Nevertheless this gives some idea of the error we might be making.

l. 15979/15 As argued above it would actually make sense to dedicate a sensitivity study to this- as this assumption (no influence on soil pools) doesn't seem very correct.

A two layer compensation approach would better be able to address this point (e.g., Nemitz et al., 2001) but this will have to be left to a future study. We believe this will certainly impact the soil nitrogen pools, but will likely not have much influence on the TAN pools modeled here. These pools are implicitly assumed to lie on top of the soil as modeled here. We will clarify this in the revised version.

l. 15980 l. 8 I think this needs some further exploration as to the underlying reasons. Can the reason be related to different management practices related to synthetic fertilizer?

In our parameterization volatilization competes with runoff and incorporation into the soil pool. This competition is evidently not significantly impacted by the rate of fertilizer breakdown. As discussed above laboratory measurements of volatilization are apparently greatly impacted by soil chemistry and pH changes in the soil. These are not included in our parameterization. We will add some further discussion here.

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