

We would like to thank the reviewer for a thorough review of this paper. The comments have helped to improve the manuscript.

GENERAL Global models need emission inventories. The problem is that it is difficult to obtain all information needed to make them. One has always to make some (crude) assumptions to make such an inventory. This is not an easy task. For that reason, this article is welcomed. It is of course also easy to criticize such work as one always can find some special examples for a country, which give other results. I hope that I have interpreted the paper correctly, as it is sometimes difficult to read. As far as I can find out from the paper, the authors are bypassing the use of animal housings and storage systems. They argue that the emission factors for spreading are not significantly different than for housing+storage (p. 15958, line 11). As far as I can find out from this formulation the authors take the emission after spreading as being representative for the whole system housing -> storage -> spreading. This is, however, not true as e.g. the emission of housing + storage is of the same order as for spreading and belongs to the SAME amount of manure. For that reason, as far as I can see, the emission should be about twice as high as the authors calculate. Moreover, they do not differentiate between spreading of manure and grazing/being in a feedyard. There are large differences in emission factors for broad spreading and grazing, which they apparently are not familiar with. For the above reason, I recommend that the paper should not be accepted, although the description of emission after spreading can be useful.

The reviewer makes the point that emission factors (with very large errors themselves) for various agricultural practices are significantly different. In this first attempt at constructing a global model for agricultural ammonia emissions suitable for use in a global earth system model (including both climate changes and biogeochemistry) we made the explicit decision to avoid explicit consideration of specific agricultural practices. As the first reviewer, Frank Dentener, notes: “*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.*” The global emissions obtained here are within the range of other published inventories. At this stage we believe it will be useful to the community at large for this model to be published. The resulting model is a first step in introducing climate dependent agricultural emissions into a biogeochemical earth system model. This should serve as a basis for future improvements both by us and other interested groups. At this point we know of no earth system model that includes climate dependent emissions. The framework described allows at least a first approximation of the impact of climate dependent emissions and biogeochemistry. Otherwise future projections will again make the implicit assumption that the impact of climate is zero.

However, we do not want to minimize the importance of various agricultural practices and feel these should be included more explicitly in refinements to the model given here. As requested by the reviewer we will discuss the issue of housing and storage in more detail, both in our discussion of future plans but also in discussing the model assumptions. Incorporating agricultural practices more explicitly is probably the top

priority in constructing a new model version in the future. This will of course be challenging on a global level as practices in Europe, for example, where emission factors are well constrained, may be significantly different than those in much of the world.

As discussed in detail below we do not believe that a neglect of animal housing would increase our global manure emissions by a factor of two.

The units for all variables in the equations should be given (otherwise, it is difficult to check whether the equations are correct or sometimes how a Henry's law coefficient is defined).

Thank you for the suggestion. We will include these.

DETAILED COMMENTS Title: should also contain the words ruminants and mineral fertilizer?

Thanks. This is probably a matter of style here. We are currently in favor of the shorter title.

Maybe N_r could be defined in the beginning. p. 15953 line 9:

Thank you, we will change.

I am not sure that Paulot et al. (2014) have derived emission factors explicitly as a function of temperature.

Temperature is implicitly included in the crop specific fertilizer application function (equation A3) and for manure (equation A6).

p. 15954 line 23: "relation between" is written here twice.

Thank you.

p. 15955 and Fig. 1: the model. Remark: It is much better to have a model where all processes are taken into account and for which is then possible to make checks that the mass balance is kept.

We make sure that the mass balance within the model domain is correctly accounted for by tracking all inputs and outputs of nitrogen to the domain considered. To fully include "all processes" would also be to include atmospheric processing and the ocean emissions of ammonia, something clearly beyond the scope of this study.

Fig. 1 (model). The fate of N_r emitted to the atmosphere is described by the CAM-chem model. If the model grid is large enough (a few hundred km's) it could be assumed that the amount of NH_3 that is emitted is deposited in this area (mainly as dry deposition of

NH₃ and wet deposition of NH_x as the atmospheric residence time is of the order of one day.

This is an important scientific question and there is no perfect solution just yet. While it is true that a lot of ammonia is deposited quickly an important fraction escapes to the atmosphere to form aerosols. This has been modeled in the atmospheric chemistry community for years using standard approaches where ammonia emissions are simply specified at the lower model boundary. The emissions simulated here (after accounting for canopy capture) are similar to those used in atmospheric chemistry models. While certainly not perfect the resulting ammonia concentrations in the atmospheric chemistry models produce suitable aerosol concentrations as well as wet deposition fields (e.g., Dentener et al., 2006; Hauglustaine et al., 2014). Nevertheless, improvements in more precisely capturing the ammonia emissions would be beneficial. A compensation point approach deals with this to some extent but the issue of sub-grid variability of NH₃ concentrations remains a problem. Another approach is having landscape Nr transfer models (Drouet et al. 2012, for example) that could eventually help in categorizing certain pfts or ecosystem types. At present earth system biogeochemistry models generally rely on reactive nitrogen fields input to the model from prior simulations. The object here is to couple the emissions and deposition in one simulation. We have also discussed this issue in the reply to the first review.

p. 15958. line 28. Why is it that only ruminants are chosen? (Potter et al., 2010 include also the excretion from pigs and poultry). Give the error made in the emissions because of this choice.

This is in error. The input includes all manure as specified from Potter et al. (2010) including that from pigs and poultry. We will correct in the text.

p. 15958. line 9. It is assumed that manure is continuously spread onto fields, bypassing the use of housings and storage facilities. It is then mentioned that the emission factors for NH₃ emissions from spreading are not significantly different from them from housing and storage (I guess this must be housing plus storage) and that for that reason the emission after spreading is used instead. This need to be discussed into more detail and I do not think that this assumption can be justified and this is crucial for the method. Indeed the emission from housings + storage facilities can be of the same order as after spreading (in e.g. kg NH₃/kg manure), but the important thing is both emissions belong to the same amount of manure (the manure is first deposited in the housing, is and subsequently transferred to the storage facility, and is then is being spread). If the emissions from housing + storage and the emissions from spreading were equal, the total emission from the whole system would almost be twice as large as the emission from spreading alone, and, as far as I can see, this is not taken into account and will lead to an underestimation of the calculated emission by a factor of two. An example: let us assume that a fraction of 0.2 of the 1 kg of N in manure entering the housing is emitted in housing (+ storage). Then $0.2 \cdot 1 = 0.2$ kg N has been emitted. Then 0.8 kg N is left when the same manure is spread. Let us assume that again a

*fraction of 0.2 of N present is emitted after spreading (= 0.2*0.8 = 0.16 kg). Then 0.2 + 0.16 = 0.36 kg of the N originally entering the housing is emitted. Therefore, although the fraction emitted is about the same for housing (+storage) as after spreading, the total emission is almost twice as high.*

We agree with the reviewer that characterizing emissions from housing is extremely important although perhaps globally daunting. We also agree that considerably more discussion is needed about the implications for neglecting these emissions. This will be included in the revised version, where we will also stress the importance of more accurately specifying agricultural emissions in future plans.

We do not believe the current method underestimates the global emissions by a factor of two. Beusen et al. (2008) updated the Bouwman et al. (1997) emission inventory to include the mass flow approach accounting for storage and housing. We note that the global emission factor derived from this approach is 19% [range 15-23%]. We have derived a global emission factor of 17%. Thus this overall emission factor derived here is rather close to that of Beusen et al. (2008). The overall emissions estimated here also agree quite well with the Paulot et al. (2014) inventory that are optimized using ammonia wet deposition fluxes.

According to the global estimate of Beusen et al. (2008) only 42% of manure is collected in animal houses and storage systems (They estimate 66% of manure is generated in mixed and landless systems. Of this 66%, 62% is collected in animal houses and storage systems for 41% of the total manure generated. Similarly another 1% of manure generated in pastoral systems is collected in storage and housing). Using the emission factors from the reviewer (EF=0.2) this would suggest total emissions of manure (M) are as follows where housing and storage is abbreviated (H&S):

$$\begin{aligned} & 0.58*EF*M \quad (\text{emissions not from H\&S}) \\ & 0.42*EF*M \quad (\text{emissions from H\&S}) \\ & +0.42*.8*EF*M \quad (\text{emissions from spreading after H\&S, where we assume .8 of nitrogen remains}) \\ & = 1.34 EF*M \end{aligned}$$

According to these emission factors this suggests by omitting housing and storage, we are underestimating the global emissions by 25% [(1-1.34)/1.34]. A number of factors may compensate for this shortfall: we are assuming no loss of manure outside of the system, and we assume emissions are not reduced due to incorporation of manure into soil.

However, there is another issue here. That is, if the manure does not pass through housing and storage it is out in the field longer and so is likely to emit more.

Consider the following analysis.

Let t_1 be the amount of time manure is within housing and storage facilities

Let t_2 be the amount of time manure is emitting after spreading

Let τ_1 be the timescale for emission from housing and storage

Let τ_2 be the timescale for emission after spreading

Let N be the amount of N in housing and storage or spread onto the fields

Thus

$dN/dt = (-1/\tau_1) N$ for nitrogen in housing and storage (assuming simple linear loss)

$dN/dt = (-1/\tau_2) N$ for nitrogen on the field (assuming simple linear loss)

So for a given initial batch of nitrogen (N_0) the amount remaining after housing and storage followed by spreading ($N_1(t_1 + t_2)$) is:

$$N_1(t_1 + t_2) = N_0 \exp(-t_1/\tau_1) \exp(-t_2/\tau_2)$$

For the same amount simply put into the fields initially ($N_2(t_1 + t_2)$):

$$N_2(t_1 + t_2) = N_0 \exp(-(t_1+t_2)/\tau_2)$$

If we assume the loss due to spreading is fixed at 20% and that due to housing and storage followed by spreading is 36% ($1-.8*.8$) then

$$[N_2(t_1 + t_2)]/[N_1(t_1 + t_2)] = .8/.64.$$

(For N_1 , 20% is initially lost in housing and storage followed by 20% of that lost in spreading so that 0.64 of the original fraction remains ($.8 \cdot .8$); for N_2 the assumption by the reviewer is that only 20% is lost so that .8 of the original fraction remains.)

This then gives a relation for the timescale of the losses:

$$[N_2(t_1 + t_2)]/[N_1(t_1 + t_2)] = .8/.64 = [N_0 \exp(-(t_1+t_2)/\tau_2)]/[N_0 \exp(-t_1/\tau_1) \exp(-t_2/\tau_2)]$$

Or

$$.8/.64 = \exp(t_1/\tau_1 - t_1/\tau_2)$$

If we assume the time for housing and storage is 45 days prior to spreading (approximately half the summer season) and τ_2 is 100 days (for the timescale for emissions following spreading, approximately the summertime timescale derived in our parameterization) then we would have the emission timescale for manure from spreading is about 50% larger than that for storage (i.e., that emissions from housing and storage is much faster than that from spreading). Velthof et al. (2012) suggests 46 Gg N ammonia emissions for 269 Gg TAN in housing and storage and 33 Gg N in ammonia emissions for 171 Gg TAN spread. Thus he finds the emission timescale from spreading is faster than that for storage, the opposite of our result.

In summary, while we neglect housing and storage, manure is left out on the field longer, compensating for the neglect of housing and storage. This of course is not a perfect compensation, but we argue that both globally and for the individual practices the error is not as egregious as suggested by the reviewer. Housing and storage will be important in a refinement of this model and will be discussed in greater detail in the revised paper.

Especially in Europe emission from storage facilities and after spreading have been reduced, making the emission from housings relatively more important. The temperature regime and ventilation regime in housings and storage facilities are also different from that on open land, leading to another emission behaviour. Moreover, processes as leaching do not take place in housings and hopefully not in storage facilities. When talking about ruminants grazing is important and that is not addressed here, although later in the article is referred to some experiments where emission during grazing is measured. During grazing most of the TAN is in the urine and urine is entering the soil at a larger speed than e.g slurry (mixture of faeces and uring). For that reason, the emission during grazing is usually much lower than during broad spreading (without using any reduction technique). See e.g. Hutchings, N.J. et al. Atmos. Environ. 35, 1959-1968.

We agree with the reviewer that it is important to emphasize the importance of regional practices. We will include a more detailed description of housing and storage and differences between grazing and spreading in our conclusions when we discuss next steps. We also think it is also important to discuss these differences earlier in the paper when we discuss our approach. We think we have provided a first step towards a climate dependent biogeochemical model of ammonia emissions and a first global estimate of the climate dependence of the ammonia emissions. New studies will provide important refinements on these initial estimates.

p. 15958. line 11: Manure is not excreted in the storage facility, only in the housing.

Thank you, we will correct.

p. 15959. Potter et al. (2010) give in their publication the N produced in manure by all domestic animals. Why are the calculations in this publication only for ruminants?

Thank you, we will correct.

p. 15959. line 11: Use import from other areas instead of lateral transport.

Thank you, we will correct.

p. 15959. line 27: It is assumed that a fraction of 0.5 of the nitrogen excreted is urine and is directly available to the TAN. Data for Europe (EMEP/EEA (2009) EMEP/EEA emission inventory guidebook 2009, Animal husbandry and manure management.) indicate that this fraction should be somewhat higher, of the order of 0.6.

Thank you for this update, but this factor is certainly highly uncertain globally. We will keep the factor of 0.5 in our model for now but will consider revising at a later date.

p. 15960. If I look at equation (2) and (5) I can see that N_resistant is transformed to N_TAN, but at p. 15960 line 9 it is stated that N_resistant is resistant to forming TAN. This should be made clearer (not everybody is an agricultural scientist and this can be confusing).

By N resistant we meant that it forms TAN slowly. We will clarify.

p. 15960. It is not clear to me what happens with the N that is subject to mechanical loss. Does the model some bookkeeping of this? (without tracking this it is impossible to have a check on the mass balance).

This N is added to the soil pools. We will clarify in the text.

p. 15964, section 2.2.6 Equations should be given for Ra and Rb. I guess that the friction velocity is part of the equation. The question is then: how is the friction velocity derived for different types of vegetation?. No information is given on that. It is mentioned "We compute average values of Ra and Rb for each CLM soil column". It should be mentioned what is done here. It is not clear to me, e.g. if every vegetation type has its own NH3 (g) concentration or not. It is e.g. not clear if first the Ra values are averaged and then the Rb values. What should be done is averaging the fluxes, not Ra and Rb values.

The aerodynamic resistance parameters are computed within CLM for individual plant functional types (PFTs) and we use these CLM parameters in our calculations. This can be found in descriptions of the CLM so we did not include the equations for these parameters. Ideally the parameters would be used exactly as the reviewer describes. Unfortunately, several important physical quantities that we use in our N pathways model are only available on the soil column level within the CLM (soil nitrogen, soil temperature, soil water etc). There can be several PFTs for each soil column so the aerodynamic resistance parameters can be averaged up to soil column level, but the ground temperature, soil water content, etc. cannot be interpolated to the finer, PFT-level grid. Therefore we compute NH3 emissions on the soil column level for which we have all the information we need. This does mean that in some columns there will be tree PFTs included implicitly in the application of manure or fertilizer. However, we believe that the impact of this contamination over large spatial scales is likely to be small since the regions of major N application are generally not PFT diverse and include mainly, or only, grass and crop PFTs.

It is mentioned that a low atmospheric concentration of 0.3 microgram/m³ is adopted, but that does not play a role as the NH3(g) concentration is usually very large. This statement is, however, not completely true. In the two or three-dimensional world the concentrations downwind are rather high, which leads to a somewhat lower emission rate as one would expect. It is mentioned that the NH3 concentration in the

future will be calculated with the CAM-model. This is, however, not so simple as it might look like. First, the vertical resolution of such a model should be very high in order to calculate near ground NH₃ concentration correctly, or other methods should be used to model the vertical concentration profile implicitly. Moreover, one should realize that concentrations in agricultural areas and nature areas within one grid element are different. It is stated that it will be assumed that f_{capture} is set to 0.6 in the future. This part, however, is not described in Fig. 1, as it is not part of the model presented here. So maybe leave out, or at least state that it is not part of the model discussed here. In order to be consequent, one should not set this to a constant factor. The factor 0.6, however, is not constant at all and depends on many factors, e.g. also on the size of the grid element used in the model. (see e.g. Asman, W.A.H. (1998) Atmos. Environ. 32, 415-421). The dry deposition of NH₃ should be modelled in the same way as the emission is, e.g. from R_a , R_b , a surface concentration etc. In addition, here it should be taken into account that different PFTs exist within one grid element. If the dry deposition of NH₃ is discussed here, it could be useful to mention that one of the removal pathways is through wet deposition (of $\text{NH}_x = \text{NH}_3 + \text{NH}_4$).

We would like to thank the reviewer for raising these points. (i) One of our next steps is to couple the emission model as described here to an atmospheric model. In the latter model the atmospheric concentration will not be constant. Dingenen et al. (2009) used a resistance analogy to estimate ozone at crop height. A similar approach can be applied to ammonia if necessary. It will be difficult to resolve different atmospheric concentrations between different PFTs, although sensitivity tests to different resolutions can be run. (ii) We agree that the factor 0.6 is not at all constant, but the canopy capture should be more explicitly implemented along with a bidirectional flux scheme when the atmospheric model is coupled. We mention this in regard to Figure 1. (iii) Yes, wet removal is important for the removal of ammonia. We will mention this in the revised paper.

p. 15966. It could be that eq. (12) is not correct. It looks like there is a K_H too much in the denominator. See e.g. Générmont and Cellier (1997), Agric. For. Meteorol. 88, 145-167. The equation depends also on how K_H is defined.

The units are given in the revised version.

p. 15968. line 24. It could maybe nice to get some information on the assumptions made by Holland et al. (2005). It looks like it is only data set. A cow in 1850 is not producing as much N as a cow in 2015. Is that taken into account by Holland et al. (2005)?

This is described in more detail in:

ftp://daac.ornl.gov/data/global_climate/global_N_cycle/comp/global_N_perturbations.pdf. These type of estimates of course are highly uncertain. It would be helpful if the reviewer could supply data or citations to support any trend in N production per cow between 1850 and present. Have the cows changed physiology, efficiency or are they producing urine or manure at different rates or is it a matter of feed?

p. 15970. The article it is assumed (p. 15958. line 9). It is assumed that manure is continuously spread onto fields, bypassing the use of housings and storage facilities. In section 3.1 the model is compared with measurements, but none of these measurements refer to emission after spreading. It are measurements during grazing and from feedyards. So this data cannot be used to test the model. Data after spreading of manure can be found in Sogaard et al. (2002) *Atmos. Environ* 36, 3309-3319. There are more data obtained since then. Sogaard et al. (2002) also indicated that other factors such as wind speed were important in Europe. It was e.g. shown that the emission rates in northern Europe were as high as in southern Europe. The effect of increase of the emission rate due to higher temperatures in southern Europe was apparently compensated for by the lower wind speeds in southern Europe. So if possible more factors should be taken into account. The same type of effect can be expected for fertilizer.

As the reviewer notes we have used studies reporting NH₃ emissions from different situations where manure is applied to the soil. This approach was used because the practice of using ‘fresh’ manure fertilizer on fields is the primary agricultural practice in countries such as India, Africa and China. We compared our modeled emissions with the grazing studies in the first instance as these studies would give us NH₃ emissions from ‘fresh’ manure which would not have lost NH₃ during storage and transport. However, we do agree if a valid comparison can be made, we can include data from Sogaard et al. (2002) in our figure for comparison. Future iterations of this model aims to include manure management processes that would give NH₃ emissions more suited to comparison with Sogaard et al. (2002). Wind speed is accounted for in determining the model emissions.

Fig. 2: Busink must be Bussink.

Thank you.

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