We thank the reviewers for their useful and insightful comments. Here we outline our responses in blue.

## Referee #1

The paper presents an extensive and well detailed model for NH3 emissions from chickens. As mentioned in the manuscript, most of the current global emission inventories for livestock are based on emission factors without any consideration for regional climate conditions and farming practices. The introduction of a few regional dependent parameters should greatly improve the spatial and temporal variations in the NH3 emissions which is of great value in for example in air quality modelling. The manuscript is well written, structured and easy to read.

## Major comments

1. Hourly and Daily timescales; the authors describe in section 2.1 that their model operates at an hourly timescale for outdoors emissions while only at a daily level for in- door emissions. While variations in temperatures inside can reasonably be expected to be small, emissions will show some variations as a function of the inside temperature, which will lead to variations in the emissions to the outside. If possible add a sentence about the choice for two different timescales and the potential impact.

We agree with the reviewer that variations in the indoor temperature of the houses will lead to variations in the emissions. The reason why the housing simulation was calculated at daily timescales is because the generalised relationship between indoor and natural temperature was derived from daily measurement data, and we want to keep the modelling structure consistent. Since the inside temperature is controlled, with variations typically smaller than diurnal variations of the outside temperature, we think simulating housing emissions with daily resolution can provide reasonable outcomes. In contrast, simulations of emissions from land spreading and backyard chickens were run with hourly timesteps in order to replicate the meteorological effects to capture diurnal variations. We propose to add a sentence in the methodology (Section 2.4.2 Global upscaling under Section 2.4 Global applications) to address this point.

2. Section 3.1.3; line 17. I would argue that the model does an average job at capturing the overall level of emissions but that some of the major changes at the start of the measurement period seem to be over and underestimated by up to a factor 2 (figure 5 May-June and ~ September). Add some discussion on the main cause for the discrepancy between the modeled and measured emissions.

We thank the reviewer for pointing out the discrepancy between modelled emissions and measurements in Fig 5. The model overestimated NH<sub>3</sub> emissions from early April to early July and then underestimated the emissions in September for House B. Following the suggestion of the reviewer, we propose to add a sentence noting this in the Results (Section 3.1.3 Resistance within chicken houses and site simulations). We know the discrepancies are mainly caused by the use of a fixed housing resistance, R\*. In reality, R\* will vary with the environmental conditions within chicken houses. However, we consider it well-justified to use a constant value of R\* in order to keep simple the overall fit of the dataset to the measured emissions, which also simplifies the global application. We agree that the value of R\* and its variation across chicken house designs is a significant source of uncertainty in our results. For this reason, we have given attention to discussing the model uncertainty related to the R\* value in the revised manuscript.

3. Uncertainties; the various uncertainties within the model are discussed to great extent but what is missing is a final summary and overall estimate of uncertainty. If possible add a table summarizing the various errors and uncertainties, including an expected (back of the envelope) range of uncertainty for each individual error. Similarly add a summary/discussion on the total expected uncertainty, and a summary for the uncertainties in the spatial and temporal distributions (similar to the ranges, at a back of the envelope level).

We thank the reviewer for this invaluable comment. We agree that there is substantial uncertainty in modelling NH<sub>3</sub> emission from livestock farming. Here, we focus on discussing the uncertainty related to model parameterizations. As stated by the reviewer, it is helpful to include a "back of the envelope" calculation of the overall uncertainty and uncertainties for individual components. The model parameters may influence the emissions interactively with non-linear consequences. We find that it is probably impossible to estimate the error based on mathematical approaches because the uncertainty distribution for many of the model terms is not well known. Instead, we conduct sensitivity analysis by simulating the effect of changes in parameters on NH<sub>3</sub> emissions. By doing this, we are able to indicate the ranges of uncertainty

and also to highlight which parameters are most important and need to be further investigated. Based on prior test, we find that indoor resistance R\*, manure pH, runoff coefficient and amount of N excreted are most important and examine these in the sensitivity tests. In addition, the uncertainty arising from the parameterization of UA hydrolysis is represented by the differences between Fig. 8 and Fig. S9. Uncertainty related to human management and processes that are not included in the model are not quantitatively investigated here but has been discussed in the manuscript.

It is worth noting that the ranges of the parameters are arbitrarily selected based on expert judgement. Indoor resistance and runoff coefficient are considered to be uncertain by a factor of 2, with manure pH uncertain by a factor ±1, which corresponds to a factor of 10x for hydrogen ions concentration. The nitrogen excretion rate is considered to have an uncertainty of 10 %. The global simulation of housing driven by varying indoor resistance values shows that 2x higher R\* leads to NH<sub>3</sub> emission decrease by approximately 31 %, and 2x lower R\* leads to 27 % higher emissions, which is similar to the result of sensitivity test at the site scale. The R\* values directly influence the magnitude of housing emissions, but only to a limited extent (as discussed in the manuscript, the primary limiting factor is the hydrolysis rate of UA). The R\* values also impact NH<sub>3</sub> emissions from land spreading of chicken manure by limit the available amount of nitrogen that is applied to land. In total, doubling R\* leads to a reduction of NH<sub>3</sub> emissions by 6.4 %, and half R\* leads to an increase of emissions by 8.5 %. The manure (system) pH, which affects the hydrolysis rate of UA and the chemical equilibria between NH<sub>4</sub><sup>+</sup> and gaseous NH<sub>3</sub>, is found to have positive effect on NH<sub>3</sub> emissions that emissions tend to increase as pH increases. We find that increasing pH from 8.5 to 9.5 causes annual NH<sub>3</sub> emission to increase by 5.8 %, while a decrease of pH to 7.5 leads to a decline of emission by 15.9 %. As the model is not able to simulate soil pH, the sensitivity analysis is carried out by changing the manure pH. The runoff coefficient was set to be 1 % mm<sup>-1</sup> for nitrogen pools in the model (Riddick et al., 2017). By doubling the runoff coefficient, the NH<sub>3</sub> emissions decrease by 11.8 %, while decreasing the coefficient to half lead to emissions increase by 16.5 %. It should be noted that among these parameters, changing the system pH has influences on both housing emissions (from broiler and layer housing) and outdoor emissions (spreading of broiler and layer manure; backyard chicken manure). The runoff coefficient only affects the outdoor emissions, while indoor resistances limit housing emissions directly, but also have impacts on consequent outdoor emissions. Smaller NH3 emissions from housing indicate a larger potential for outdoor release during the spreading stages under the same farming

practices. Conversely, higher housing emissions lead to smaller emission potential for land application because "what has been emitted is not going to be emitted again". Uncertainty related to the runoff coefficient has both spatial and temporal variations, which is because regions and periods with higher precipitation are more influenced than dry areas and periods. Concerning the nitrogen excretion rate from chicken, find that a 10 % of variation leads to an annual NH<sub>3</sub> emission change of approximately 12 %. The change in NH<sub>3</sub> emission is not proportional to the nitrogen input because of non-linear interactions in the model, e.g., an increase in nitrogen input by 10 % may only lead NH<sub>3</sub> emissions to increase by a negligible amount in regions with heavy rainfall.

Combining these ranges and taking the base run result as the "best estimate", the overall uncertainty is estimated as the square root of the sum of the squares of the individual uncertainties, expressed as mean values of magnitudes of positive and negative changes from the sensitivity tests. For housing emissions, the estimated uncertainty is 33 %, which combines uncertainty from indoor resistance on housing emissions (29 %), manure pH (11 %) and excreted nitrogen (12 %). The uncertainty of emissions from chicken manure land spreading is 18 %, resulting from uncertainty in manure pH (11 %) and runoff coefficient (14 %). The uncertainty of emissions from backyard chicken is 21 %, which combines uncertainty from excreted nitrogen (12 %), manure pH (11 %) and runoff coefficient (14 %). The total expected uncertainty in annual global emissions of NH<sub>3</sub> is estimated to be 22 % of the total global emissions, corresponding to 1.2 Tg N per year. This value is determined by combining all component uncertainties, i.e. indoor resistance for emissions from both housing and land spreading (together 7 %), manure pH (11 %), runoff coefficient (14 %) and excreted nitrogen (12 %), assuming that they are independent. We summarize these results in Tables R.1.1 and R1.2 below, which we propose to include in the revised manuscript in response to the comment from the reviewer.

Table R1.1 Sensitivity test for model parameters for global application of the model.

Parameter	Value tested	Value change	ΔNH <sub>3</sub> em	ission %
	16700 s m <sup>-1</sup> (base)	1 x	0.0 %	
<sup>a, b</sup> Indoor resistance, R*	8350 s m <sup>-1</sup>	0.5 x	<sup>a</sup> 27.1 %	a, b 8.5 %
	33400 s m <sup>-1</sup>	2 x	a -30.6 %	a, b -6.4 %

	8.5 (base)	8.5 (base) 1 x 0	
a, b, c Manure pH (H <sup>+</sup> )	7.5	0.1 x	-15.9 %
	9.5	10 x	5.8 %
b, c Runoff coefficient, Rrunoff	1 % mm <sup>-1</sup> (base)	1 x	0.0 %
	0.5 % mm <sup>-1</sup>	0.5 x	16.5 %
	2 % mm <sup>-1</sup>	2 x	-11.8 %
	11.2 Tg N year <sup>-1</sup> (base)	1 x	0.0 %
a, b, c Excreted nitrogen	10.1 Tg N year <sup>-1</sup>	0.9 x	-12.3 %
	12.3 Tg N year-1	1.1 x	12.6 %

<sup>&</sup>lt;sup>a</sup> Parameters affect NH<sub>3</sub> emissions from housing. <sup>b</sup> Parameters affect NH<sub>3</sub> emissions from land spreading of chicken manure. <sup>c</sup> Parameters affect NH<sub>3</sub> emissions from backyard chicken.

Table R1.2 (manuscript Table 1) Excreted nitrogen from housed and backyard chicken, and estimated annual NH<sub>3</sub> emissions from each practice based on 2010

Production system	Total excreted nitrogen (Tg N)	Practice	Total emission (Tg N)	Average P <sub>V</sub> (%)
Broiler and	9.0 [±0.9]	Housing	2.0 [±0.6]	22 [±7] %
layer	310 [=013]	Land spreading	2.7 [±0.5]	39 [±7]* %
Backyard chicken	2.2 [±0.2]	Left on land	0.7 [±0.2]	32 [±7] %
Total	11.2 [±1.1]		5.5 [±1.2]	49 [±11] %

<sup>\*</sup> Based on the excreted N remaining (i.e., 7.0 Tg N) after NH<sub>3</sub> volatilization from housing.

4. Current inventories; that brings us to a comparison to current inventories which is as of yet missing in the manuscript. Most regional/country scale inventories, to some extent, do have emission totals for chicken housing/open-range chickens. How do the emissions reported in this manuscript compare to some of those emission inventories (for example, UK, Netherlands, Denmark, US, German inventories. . .etc), and did the added complexity of the model improve the overall uncertainty in the emission totals?

We thank the reviewer for this comment. We reply to this point together with *Comment 5*, please see our answer below.

5. Similarly, add some discussion on the average Volatilization levels reported in this study compared to those in current literature.

We thank the reviewer for these comments that comparing the results with existing inventories and literature. Here we can compare the results from the AMCLIM model to three other (model-based) studies/reports from Denmark, Netherlands and United Kingdom, respectively. The Danish IDA model (Albrektsen et al., 2017) and the UK NARSES model (Misselbrook et al., 2011) provided 2010 emission data, and the NEMA model (Velthof et al., 2012) from Netherlands estimate emissions in 2009 (see Table R1.3 below). It is important to clarify that all these studies show emissions from poultry rather than chicken. It has been clearly stated that the input used in the AMCLIM from the GLEAM model used here are chicken data, which excluded other poultry such as turkeys, ducks etc. Therefore, we can see that the excreted nitrogen from the GLEAM model (GLEAM FAO, 2018) is generally smaller than other individual studies. For housing, the AMCLIM model shows similar estimates of NH<sub>3</sub> emissions to the other models. The housing emissions given by the AMCLIM model are smaller than the local models in Denmark and Netherlands, partly due to the smaller total excreted N from animals. However, the AMCLIM model suggests larger emissions from land spreading for Netherlands and the UK (spreading-derived emissions are not available from the IDA model), especially in Netherlands where the difference between the two estimates reaches 8 x. This is probably due to the different schemes or assumptions for land spreading practices, i.e. deep injection of manure, in different models. The P<sub>V</sub> rates, which indicate the fraction of nitrogen that is emitted as NH<sub>3</sub> are comparable from all models for the housing sector. The AMCLIM model suggests that the P<sub>V</sub> rates do not vary significantly between these countries because the indoor conditions are largely controlled and in similar climates, which leads to small variations in house environments. This table will be included in the revised manuscript.

It should be noted that there is a lack of published experimental data on emissions from chicken in many climate (e.g. tropical climates), for which future measurements datasets would be useful to further test the model performance. We compare the model performance with experimental field studies in answer to reviewer 2 (see Figure R2.1 in reply 2).

Table R1.3 Estimates of NH<sub>3</sub> emissions from poultry/chicken farming by IDA for Denmark (Albrektsen et al., 2017) and by NARSES (Misselbrook et al., 2011) for the United Kingdom based on 2010, and by NEMA (Velthof et al., 2012) for Netherlands based on 2009\*. Ranges given in the P<sub>V</sub>-housing represents the geographical variations across the country.

	Ammonia	Ammonia		
	emission from	emission from	Total excreted N	D. housing (0/)
	Housing (Gg N	Spreading (Gg N	$(Gg N yr^{-1})$	P <sub>V</sub> -housing (%)
	yr <sup>-1</sup> )	yr <sup>-1</sup> )		
Denmark	3.0 (IDA)	Not available	11.3 (IDA)	26.5
Denmark	1.7 (AMCLIM)	2.4 (AMCLIM)	7.9 (GLEAM)	21.5 (20.4 – 22.9)
Netherlands	11.4* (NEMA)	1.8* (NEMA)	62.9* (NEMA)	18.1*
Netherlands	10.0 (AMCLIM)	15.0 (AMCLIM)	49.0 (GLEAM)	20.4 (20.0 – 21.0)
United	15.0 (NARSES)	14.7 (NARSES)	Not available	17.8
Kingdom	17.4 (AMCLIM)	23.7 (AMCLIM)	84.1 (GLEAM)	20.7 (18.6 – 22.1)

### Minor edits and remarks

a. Figure S1, is there any reasoning behind the choice of a third order polynomial?

We use this third order polynomial equation to represent a generalised relationship between indoor and outdoor temperature because 1) it is roughly consistent with a simplified parameterization proposed by (Gyldenkærne et al., 2005) that the indoor temperature behaves in a "increase-stay-increase" pattern, 2) and it is applicable and convenient for computing. We will add this clarification to the revised manuscript in Section 3.1.1 Temperature of chicken houses.

b. Page 6., line 9, add "of" between lack and knowledge.

Corrected, thanks.

## References

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- Velthof, G. L., van Bruggen, C., Groenestein, C. M., de Haan, B. J., Hoogeveen, M. W. and Huijsmans, J. F. M.: A model for inventory of ammonia emissions from agriculture in the Netherlands, Atmos. Environ., 46, 248–255, doi:10.1016/j.atmosenv.2011.09.075, 2012.

# Referee #2

## General comments

The manuscript "A climate-dependent global model of ammonia emissions from chicken farming" from Jize Jiang et al., describes a model of ammonia volatilization from chicken farming: AMCLIM- Poultry.

The model is based on a simple approach were urea hydrolysis to ammonium and ammonia is implemented for emissions in buildings, in field applied with chicken manure and in farm backyards. A resistance approach is used and specific resistance parameterisation is used for buildings. A simple mass balance approach is used to treat manure water content.

The model is compared to measurements in a few US farms and applied to evaluate worldwide emissions from chicken farming, based on FAO statistics.

The issue is of great interest for the scientific community as ammonia emission is a key component of air quality prediction and environmental impacts and emissions from chicken farming is still not well developed. The presented study is based on the work of Elliot and Collins (1982) for hydrolysis and combined with a resistance approach. The application of the model at the global scale is of great interest, and especially the analysis of the humidity and temperature dependent NH3 emissions as well as the dataset constructed for that purpose.

This manuscript should be published provided some the authors answer some comments on the model design.

• Model: The model is key in this manuscript and it is both very simple but it accounts for the most important processes about the environmental conditions, which makes it effectively very useful. The presentation of the model may however be improved by first exposing clearly, right at the beginning, the hypothesis behind it, second condensing the description in the material and methods only, whereas it is now split between sections, and third, better explicating the model for manure spreading in the field.

We thank the reviewer for these constructive and insightful comments; our reply is listed in detail below.

o Regarding model hypothesis, I found several hypotheses that were not always explicit: i) there is no transfer resistance in the litter itself (eq. 7); ii) ammonium is considered the only form of TAN in the liquid phase (eq. 6); iii) the pH is considered not influenced by the UA hydrolysis; iv) NH4+ is consider to be completely free in the litter and soil and not to be bound to soil or litter particles; v) the system is considered to be litter only but no soil; vi) No exports are in the equations but the model is initialised at each house cleaning; vii) there no litter evaporation is considered in the houses, rather an equilibrium is considered.

We thank the reviewer for pointing out that these hypotheses need to be explicitly described in the manuscript. We will update the manuscript to include briefly the following points in the methods section (according to the numbering used above by the reviewer):

- i) There is no explicit term for transfer resistance in the litter that is simulated in the model. Instead, the housing resistance R\* is considered to include an "integrated" resistance that consists of aerodynamic and boundary layer resistances and also the resistance of litter.
- ii) In the model version used in the initially submitted manuscript, we considered that aqueous TAN is mainly in the form of  $NH_4^+$ . For the revised manuscript, we have now improved the model by including the dissociation constant for  $NH_4^+$  ( $K_{NH4}$ ) and generalise the Eq.6 as follows,

$$\Gamma = \frac{[NH_4^+]}{[H^+]} = \frac{[TAN]}{K_{NH_4^+} + [H^+]} = \frac{M_{TAN}}{V_{H_2O}(K_{NH_4^+} + [H^+])}$$

iii) We used a fixed pH of 8.5 rather than including a dynamical scheme for determining the pH. We appreciate that pH increases as UA hydrolyses, which causes larger instantaneous NH<sub>3</sub> emissions, similar to the effect simulated for urea by Móring et al. (2016). However, such an approach substantially complicates the model and involves substantial additional unknowns. For a practical model targeted for global upscaling, we therefore consider this simplification appropriate. We find that the changing the pH of the manure by ±1 causes the annual NH<sub>3</sub> emission to change by -15.9 % to 5.8 %. While the time course of instantaneous emissions changes, the uncertainty in the annual emission is smaller than the instantaneous effect, as this is constrained by the total amount of UA hydrolysed.

- iv) We simplified soil processes when simulating NH<sub>3</sub> volatilization from manure spreading. The volatilization of NH<sub>3</sub> is considered to be a much quicker process compared to the immobilization of NH<sub>4</sub><sup>+</sup> in the soil. In addition, the adsorption of TAN to soil is not simulated in this model because it requires detailed soil chemistry which is only achievable by using more detailed land models. This could be a future direction of study, also considering the effect of manure incorporation into the soil.
- v) We considered that manure or litter is the major substrate of TAN. This can be true because 1) there is no soil in chicken houses and 2) chicken excretion is relatively dry and with large fraction of solid materials compared to other livestock. The model thus cannot simulate interactions between manure and soil, after spreading. As mentioned above, this could be a potential future area of model development.
- vi) We do not include an export term in the mass balance equations. Instead, we set each pool to zero when there is an emptying event. The assumption is that when the houses are cleared out, this is complete, and all the cleared manure ends up being spread on local fields under current model resolution of  $0.5 \times 0.5$  degree.
- vii) We do not simulate litter evaporation explicitly in houses because the model for housing simulation is run at daily time basis. The chicken excretion is relatively dry, and we assumed there is no extra water added to the system. It is a simplification that the manure has equilibrium moisture content after a day. The uncertainty has been discussed in the manuscript.

As requested by the reviewer, we will update the manuscript and clearly state the points above in the methods section.

o Regarding the description of the model, it would be much easier to read if the whole model could be defined at once in the material and methods: factors affecting UA hydrolysis should be presented in the material and methods. Watch out that the TAN is sum of NH3l+ NH4 and you should justify NH4 >> NH3.

We will update the manuscript to present the factors affecting UA hydrolysis in the Methods section. While we agree that  $[NH_4^+] >> [NH_3]$  we have now also updated Eq.6 (see point ii above) to better simulate the partition between  $NH_3$  and  $NH_4^+$ .

o Regarding the manure spreading, it is unclear how VH2O is calculated in this situation, and the description of run off is quite unclear.

The  $V_{H_2O}$  in outdoor simulations (manure spreading + backyard chicken) is calculated from the mass of water in the system,  $M_{H_2O}$ , from Eq. 14. The runoff is determined from a runoff coefficient multiplied by the amount of water that is available for runoff, which is determined by subtracting the water absorbed by the manure from the rainfall. We will update the manuscript to make this explicit.

• UA hydrolysis fitting to RH and TA: Did you try fitting on vapour pressure pvap = RH/100\*psat(Ta)? In addition, did you try fitting on both Ta and RH together?

The RH and temperature dependence of UA hydrolysis are taken from Elliott and Collins (1984) and Riddick et al. (2017). Both studies used a combined influence, which is a product of individual factors as expressed by the Eq. 20. The impact of RH on UA hydrolysis is associated with the equilibrium moisture content, which depends on temperature and RH. We do not fit on multiple variables simultaneously. Instead, we decomposed the effects from each factor to normalise the UA hydrolysis rate. We appreciate that fitting UA hydrolysis to vapour pressure as well as vapour pressure deficit could be a future investigation.

• Literature: I feel that some important papers may be lacking. In particular, on ammonia emissions data and models from land spreading manure or urea hydrolysis. The literature is much more abundant on dairy cow or pig manure, but I was wondering if and why it would not be possible to refer to these when building up the model for chicken manure. Some examples given here

We thank the reviewer for listing these useful articles. We will discuss and include relevant papers. Sigurdarson et al. (2018) presented a comprehensive review for ammonia emissions from urea hydrolysis, which implicates important mitigation measures. McQuilling and Adams (2015) developed a model for estimating NH<sub>3</sub> emission from livestock in the United States. The paper is developed from McQuilling's PhD thesis that established an emission inventory for the US including poultry. We also use the paper of Miola et al. (2014) and literature cited therein to further evaluate our model performance for field application of poultry litter.

o Ammonia Volatilization after Surface Application of Laying-Hen and Broiler-Chicken Manures. By: Miola, Ezequiel C. C.; Rochette, Philippe; Chantigny, Martin H.; et al. JOURNAL OF ENVIRONMENTAL QUALITY Volume: 43 Issue: 6 Pages: 1864-1872 Published: NOV-DEC 2014. Typos: please check thoroughly the text for typos.

o The molecular processes of urea hydrolysis in relation to ammonia emissions from agriculture By: Sigurdarson, Jens Jakob; Svane, Simon; Karring, Henrik. REVIEWS IN ENVIRONMENTAL SCIENCE AND BIO-TECHNOLOGY Volume: 17 Issue: 2 Pages: 241-258.

o Modeling and measurements of ammonia from poultry operations: Their emissions, transport, and deposition in the Chesapeake Bay By: Baker, Jordan; Battye, William H.; Robarge, Wayne; et al. SCIENCE OF THE TOTAL ENVIRONMENT Volume: 706 Article Number: 135290 Published: MAR 1 2020

o Semi-empirical process-based models for ammonia emissions from beef, swine, and poultry operations in the United States By: McQuilling, Alyssa M.; Adams, Peter J. ATMOSPHERIC ENVIRONMENT Volume: 120 Pages: 127-136 Published: NOV 2015

• Consider shortening the discussion. I found the discussion a bit long with a few redundancy and repetitions.

We will update the discussion to make it more concise.

• A comparison with existing emission factors would be very interesting

We thank the reviewer for this insightful comment. We will add a comparison with existing emission factors. In particular, we take note of the review of experiments by Moila et al. (2014) and have addressed this further for inventories below.

• Typos and English. I suggest double-checking the spelling and phrasing of the manuscript.

We thank the reviewer for this considerate suggestion, and we will update the manuscript.

Detailed comments

*P2.L17-18:* Could you be more specific on which parameters were tested?

The effect of temperature and slurry dry matter content on NH<sub>3</sub> volatilization were based on the review of Sommer and Hutchings (2001). We will mention this in the revised manuscript.

P3.Eqns (1-3): In these two equations, the export flux of excretion by removal during house cleaning is not considered. It would be clearer to add it. This would allow all Mexctretion, MUA and MTAN to get down to zero when the house is cleaned.

Agree. We set the N pools to zero when the house is cleaned and will make this explicit.

P4.L1: FTAN is not a conversion rate but a flux. Please consider revising.

Agree. We will correct and update the manuscript. We change " $F_{TAN}$  is the conversion rate of UA to TAN" to " $F_{TAN}$  is the flux of TAN that is decomposed from UA hydrolysis".

P4.L11: and eq. 4: it would be good to give expression of K here rather than in the results section.

Agree. We will move this part to the method section.

P4eq. 6 is not strictly speaking true since MTAN = MNH4+ + MNH3. Does this mean you consider MNH3 negligible compared to MNH4+? You could easily express MNH3 as a function of MNH4+ based on the dissociation constant and pH and then get a corrected expression for equation 6 that accounts for the pH.

Agree. As answered previously, we have corrected the Eq.6 to include the dissociation constant for NH<sub>4</sub><sup>+</sup>, which then allows both NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> to be included.

P4L26-27: the justification of using the same approach for backyard and field may be more developed. Especially, how the interaction with the soil is treated.

The same approach used for simulations of land spreading and backyard chicken refers to the broad resistance approach, which differs from the indoor resistance R\* method. In this study, the interaction with the soil was not simulated, which is consistent with the GUANO model described by Riddick et al. (2017) which was validated for measured NH<sub>3</sub> emissions from seabird guano. The major difference between land spreading and backyard chicken is that we incorporated crop calendar dates to determining the timing of manure application for land spreading, whereas for backyard chicken excreta is deposited to land all year. Whereas ultimate immobilization, plant uptake or nitrification of TAN in the soil are not treated (since these are typically slower processes than NH<sub>3</sub> volatilization), these loss terms can be considered

implicitly as part of the uncertainty associated with depletion of deposited excreta by run-off. We will outline these points in the revised discussion, while further assessment of these interactions offers scope for future work

P5L7-8: NH3 is removed but also fresh air dilutes NH3 in the building: both process occur.

Agree. We will rephrase and update the manuscript.

P5 eqns 8 and 9: From what I understand here, the litter (or excretions) has a humidity, which is in equilibrium with atmospheric humidity in the building (express by RH and T). This is similar to soil surface humidity that is in equilibrium with the atmosphere just above. Could-you explain the process behind equation 9?

Equation 9 is based on the hygroscopicity of poultry litter and so accounts for the moisture absorbed by the litter as it reaches an equilibrium state, which is dependent on temperature and RH. The litter moisture content exerts a vapor pressure on the adjacent air, and the ratio of this moisture vapor pressure to the saturated vapor pressure of pure water in air at the temperature of the material is called the equilibrium relative humidity (Henderson, 1976). If the air RH is higher than the equilibrium relative humidity of the material, the material will increase in moisture content. Conversely, the material will decrease in moisture content if the air RH is lower than the equilibrium. We assume that the litter moisture content instantaneously maintains equilibrium with the housing environmental temperature and humidity, which we will clarify in the revised manuscript.

P6L1: The pH should be influenced by UREA hydrolysis, isn't? Could you better justify the choice of fixing the pH?

As answered previously (by iii), we do not include a dynamical scheme for determining the pH that can be influenced by the UA hydrolysis. We choose a fixed pH value of 8.5 to represent the system pH, which is a typical value of chicken excretion pH (Elliott and Collins, 1982). This is much more practicable for a global model than attempting to simulate explicitly the dynamic pH response of litter to UA hydrolysis, which depends on poorly known buffering capacity and may also vary between microsites (Móring et al., 2016). By carrying out sensitivity tests, we find that varying pH only leads to small change in total annual NH<sub>3</sub> emissions, where increasing pH leads to larger emissions over a shorter period, while reducing

pH because leads to slower but more sustained emissions. Increasing pH from 8.5 to 9.5 cause annual NH<sub>3</sub> emission to increase by 5.8 %, and a decrease of pH to 7.5 leads to a decline of emission by 15.9 %.

*P6L28: I suggest explicitly stating that Qxout has been neglected.* 

Agree. We will explicitly state that Qxout has been neglected due to the negligible ambient concentration of NH<sub>3</sub> compared to indoor concentration. We will update the manuscript.

P6 eq 12-13: fundamentally, this equation would also hold for water in buildings: hence, humidity in the building may depend on the rate of air renewal and the surface humidity. This would mean that pvapin =  $f(pvapout, Q, R^*, pvapsurface)$  but also that there would a removal flux for humidity also. Proportional to  $Q^*(pvapin-pvapout)$ . Could you elaborate on that and justify better, why evaporation from building is neglected?

As answered previously (by vii), we do not simulate litter evaporation in houses because the model for housing simulation is run on a daily time basis. The chicken excretion is relatively dry, and we assumed there is no extra water added to the system. It is a simplification that the manure has an equilibrium moisture content after a day. The uncertainty has been discussed in the manuscript.

P7L3-4: I suggest defining clearly, what the "system" is: is it the litter only, or the litter plus a certain depth of soil?

The system refers to the manure only, and soil processes are not simulated in the model. We will clarify the system definition in the manuscript.

P7L8-9: Could you explain better why the water amount in the system could not be less than that in the excretion? Indeed, since evaporation occur, the water amount may become lower.

As mentioned previously, we assume that the litter moisture content is in equilibrium with the environment. The model precludes a dynamic evaporation simulation for the litter. The litter tends to get drier if the humidity falls, and wetter if the humidity increases. The amount of water of the system should not be less than the equilibrium moisture content of the excretion. We will update the manuscript to clarify.

P7, section 2.3: The field application is unclear and would need further details: 1) TAN in soil is known to be in equilibrium with clay, explain why this process is neglected. 2) The evaporation equations as well as the expressions of the resistances are not given and should be detailed, in the supplementary at least. 3) How is VH2O calculated in that situation?

1) As noted above, the AMCLIM model does not include an interactive scheme for TAN and soil. We consider that chicken manure is mainly lying on the surface of crop lands because it is relatively dry and is not physically mixed with underlying soils. This means that the model as presented does not consider the potential benefit of immediate incorporation of poultry litter into soil. Meanwhile, simulating the interactions with soil would require a more detailed characterization of soil chemistry, which might only be achieved by employing a sophisticated land model. Therefore, we exclude soil processes that require more detailed information of soil properties, which is beyond the capability of this model. 2) Compared to the housing simulations that use equilibrium moisture content, for simulations of land spreading and backyard chicken, we used the evaporation data from ECWMF to determine the water pool. The resistances (R<sub>a</sub> and R<sub>b</sub>) for NH<sub>3</sub> volatilization are calculated based on Seinfeld and Pandis (Seinfeld and Pandis, 2016). We will add a description of resistances in the supplementary materials. 3) As answered previously, the V<sub>H2O</sub> in outdoor simulations (manure spreading + backyard chicken) is calculated from the mass of water in the system, M<sub>H2O</sub>, from Eq. 14. The runoff is determined from a runoff coefficient multiplied by the amount of precipitation water that is the rainfall subtracts the water absorbed by the manure. We will make this explicit in the revised manuscript.

P8L28: but evaporation ay also occur in the building. Please comment.

As answered previously, we assume that the litter moisture content is in equilibrium with the housing environment. We used the equilibrium moisture content to determine the water content of the litter.

P8L30: "houses were empty in different months". Please rephrase as this is unclear what it means.

The context is as follows: "12 simulations were run by assuming that chicken houses were emptied in different months for each simulation, i.e. from January to December, and the simulations started in corresponding month." To clarify our message, we will change this as

follows in the revised version: "To calculate the varying impacts of emptying the chicken houses at different times of the year, we ran 12 different year-long simulations: each starting from a different month, i.e. from January to December, and assuming the chicken house had just been emptied."

P9eq 18: I suggest using the term Navailable instead of Nsoil\_poultry. It is also unclear from the text, whether N total includes manure and mineral nitrogen

We change the  $N_{Soil\_poultry}$  to  $N_{available}$ .  $N_{total}$  includes nitrogen from manure fertilizer, of which nitrogen from chicken manure is only a small fraction considering the model grid resolution and the spatial distribution of other sources.

P10L21-22: It is unclear when the building temperature is not used, what temperature is then used? Please clarify.

A distinction needs to be made here between: i) the derivation of relationships between inhouse and outdoor temperature for the model parametrization and ii) running of the AMCLIM model for global upscaling. The text here refers specifically to the former. In this case, the data for when broilers are <0.5 kg per bird are excluded from the parametrization because a) broilers smaller than this size do not contribute significantly to NH<sub>3</sub> emissions and b) houses are kept warmer than normal for the smallest chicks was compared with birds >0.5 kg. By excluding these data for small birds, a much better relationship can be found between indoor and outdoor temperatures (Fig. S1), which is also representative of the periods of significant NH<sub>3</sub> emissions. In running the AMCLIM model for global upscaling, the same relationship from Fig. S1 is applied for all weights of birds. This will tend to underestimate the temperature in houses for birds <0.5 kg, but as noted this will have negligible effect on total emissions, because these are dominated by periods when chicken are >0.5 kg weight. We will clarify this in updating the integrated description of the methods.

P10-P11: section 3.1.2 should be in the material and methods section and not in the results as it is a model description to me.

Agree. We will move this to the method section.

P11 eq 23: To me it would be more logical if urea hydrolysis would be dependent on the excretion humidity %me rather than RH. However, the two are linked. Could you comment on that?

As noted above, the housing model is run on a daily time-step, since this is the time-scale for which we have measured emission data for verification. This means that we need to identify a representative litter humidity for daily periods for use with the parametrized relationship between litter humidity and hydrolysis rate. Bird excreta is actually liquid, but the water will be dispersed in a litter-based system throughout the litter. If it is envisaged that fresh excreta reaches equilibrium with the surrounding litter within an hour or a few hours, then this means that for a daily simulation it is more representative to use the litter humidity in equilibrium with daily humidity data. We will add a comment to this effect in the methods.

P11-L16-17: "emissions were due to unavailable measurements": this sounds weird: could you rephrase?

For the revised manuscript we propose to change "Gaps occurred in measured NH<sub>3</sub> concentration and emissions were due to unavailable measurements, while the model was kept running." into "Gaps shown in measured concentrations and emissions of NH<sub>3</sub> represent unavailable measurements, while the model was kept running during gaps to produce continuous output."

P12 section 3.3: the model for manure spreading was not tested at all, while the model for housing was tested. Would there be any dataset to demonstrate the quality of the model for outdoor application? Alternatively, would there be any paper to refer to on that?

We will make it clear that, from an experimental perspective, the AMCLIM model builds on the approach of the GUANO model, which has been tested in a wide range of outdoor climatic conditions (Riddick et al., 2018). In addition, we propose to include a brief comparison with the studies summarized by Miola et al. (2014), based on comparison of the  $P_V$  values (i.e. % of TNA of Miola et al., % of Total N applied).

To address this, we ran a set of simple site experiments for land spreading to quantify the  $NH_3$  volatilization rates ( $P_V$ ) under different environmental conditions. We set the application rate to  $100 \text{ kg N ha}^{-1}$  (equivalent to  $10 \text{ g N m}^{-2}$ ), which is comparable to the value used in Rodhe

and Karlsson (2002) (110 kg N ha<sup>-1</sup>), Sharpe et al. (2004) (109 kg N ha<sup>-1</sup>, 99 kg N ha<sup>-1</sup>, 133 kg N ha<sup>-1</sup>) and Marshall et al. (1998) (70 kg N ha<sup>-1</sup>). The model is driven by the mean daily air temperature given from the previous studies, while the diurnal variations of temperature and other meteorological factors such RH and precipitation are not available from these publications. The ground temperature is assumed to be 2 ° C higher than the air temperature, where ground temperature is not available from the published experiment. The sum of aerodynamic and boundary layer resistances is assumed to be 100 s m<sup>-1</sup> as it cannot be calculated due to the lack of environmental inputs provided by the authors. The wash-off pathways of the model were shut down due to the unknown rainfall information, so the simulations are representative of rain free experimental conditions. We initialized the model simulation using a 7-day period prior to application of chicken litter, to allow initialisation for each nitrogen pools. The model was then run for 21 days to determine the NH<sub>3</sub> volatilization. We compare the modelling results with reported measurements from five experimental studies (Lau et al., 2008; Marshall et al., 1998; Miola et al., 2014; Rodhe and Karlsson, 2002; Sharpe et al., 2004), as shown in Fig. R2.1. We focus on experimental data for chicken that are broilers or layers (rather than other poultry, e.g. turkey) and data for "young" litter which was stored for a short period before application, normally less than a week or 10 days. There are three groups of comparisons that represent different simulation and measurement duration.

As shown in Fig. R2.1, the simulated volatilization rate of  $NH_3$  increases as temperature increases, because of the faster UA hydrolysis rate in hotter conditions. The shaded areas illustrate ranges of  $P_V$  from simulations that use different RH values ranging from 20 to 100 %, while the solid lines represent the mean  $P_V$  rate for the range of RH values for each simulation period (7, 14, 21 days).

Compared with the experimental studies shown in Fig. R2.1, the model application underestimates NH<sub>3</sub> volatilization for the 21 days simulation and overestimates for the 14 days simulation. However, it is evident that these experimental studies also show large variations, which we expect is especially due meteorological variation within and between the experimental studies, such as rainfall or windy conditions. For example, at a mean temperature of around 26 °C Sharpe et al. (2004) reported P<sub>V</sub> of 23 % and 5 %, respectively. The latter value was caused by a rain event taking place two days after application, explaining why the latter point appears low on Fig. R2.1 where the simulations are based on rain free conditions.

Overall, the model provides  $P_V$  rates that falls within the range between 0.5 x to 2 x compared to the measurements. It should be noted that this is a very simple model experiment as several features of the AMCLIM-Poultry are not available because the published experimental studies do not fully describe environmental conditions.

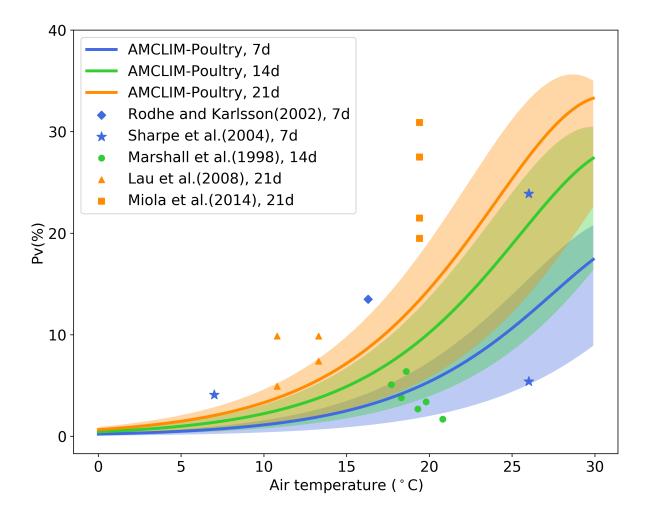


Figure R2.1 Simulated fraction of total applied nitrogen that is loss as NH<sub>3</sub>-N ( $P_V$ , %) as a function of air temperature (°C) by the AMCLIM-Poultry for simulating periods of 7, 14 and 21 days, and comparison with experimental studies that measured NH<sub>3</sub>-N loss for 7, 14 and 21 days. Simulations conducted for rain-free conditions, where shaded areas indicate the range for simulations from 20 % to 100% relative humidity. The figure of 5 % volatilization at 27 °C by Sharpe et al. (2004) was associated with high precipitation.

P14- L6-13: it is actually unclear in the previous part if the papers how RH and Ta are modelled in houses.

We used the outdoor RH to represent the indoor RH for the housing simulations because the indoor and outdoor RH were found to be comparable from the USEPA AFO's dataset. The indoor temperature was determined by using generalised relationships shown in Fig. S1 based on AFO data. We will make this clearer in the revised methods section.

P14-L14-20: I would suggest adding a table with durations, temperatures and may be RH conditions for the different chicken houses managements discussed

The environmental variables of the houses including temperature and RH vary with time. We have shown the variations in Figs. 4 and 5.

P14-15 section 4.1: it is a bit confusing here to understand how the RH-dependency of urea hydrolysis is used in outdoor conditions. Please detail.

Section 4.1 is the discussion of parameterization of housing simulation instead of outdoor simulations. We simulated NH<sub>3</sub> emissions from chicken housing by using both the RH dependency of UA hydrolysis from Elliott and Collins (1982) and that is derived from USEPA AFO's dataset. The results are shown in Fig. 8 and Fig. S9, respectively. The RH dependency of UA hydrolysis used for outdoor simulations is from Elliott and Collins (1982), which has been previously tested and found to provide robust estimates from the GUANO model (Riddick et al., 2017). We will clarify the text accordingly.

P16L1-10: the whole paragraph except last sentence is quite unclear. Please rephrase. In the last sentence, it may not be true that sensitivity is negligible though, since  $R^*$  may be very variable among situations.

We will rephrase this paragraph. We have now carried out a set of sensitivity tests for global simulations that detail how NH<sub>3</sub> emissions vary with several uncertain parameters (Table R2.1). We find that varying indoor resistance values, R\* by a factor of 2 causes NH<sub>3</sub> emissions to change by approximately 30 %: 2x higher R\* leads to NH<sub>3</sub> emission decrease by approximately 30 %, and 2x lower R\* leads to 27 % higher emissions, which is similar to the result of sensitivity test at the site scale.

P16-L27-33: Could we not say that for very large RH, since UA hydrolysis is so effective, there is a limiting effect due to the non-availability of total nitrogen in the system after a certain time?

We agree that this could happen in principle, but suggest that this cannot explain the results of our steady-state model run as summarized in Fig. 7b. Firstly, if total N were limiting, then this would mean that the value of P<sub>V</sub> would not increase further above a certain threshold. However, we see that the value of P<sub>V</sub> actually *decreases* above 80% RH, pointing to the need for a different explanation. As we have noted, with excess water available, there is a dilution effect on TAN concentration, which can explain this feature. Secondly, we would expect that total N would become limiting once all available UA is hydrolysed (equivalent to 60% volatilization rate of total excreted N). However, we do not find this threshold to be exceeded. Therefore, we consider the dilution effect to be the likely cause of this decrease in P<sub>V</sub> above 80% RH.

## P17L8-9: Difficult to understand. Please rephrase this sentence

For the revised manuscript, we propose to change "Considering the variations in  $P_V$ , there is most estimated variation in NH3 volatilization of manure spreading and backyard." into "Considering the  $P_V$ , the most significant spatial variations relate to emissions from manure spreading and backyard chicken, with less spatial variation in  $P_V$  for housed birds"

## P18L26: It is unclear why initial water in excretion is not accounted for. Please rephrase.

We explain the reason in P18L24-25, "The model is not able to simulate the evaporation from the litter in the chicken house. Therefore, the litter moisture is assumed to be at equilibrium". As answered previously (reply to comment on *P11 Eq.23*), chicken excretion is relatively dry compared with other livestock excreta, so we assumed it takes a much shorter time for chicken litter to reach equilibrium moisture content than the modelling timestep (1 day), allowing use of the equilibrium value.

P18-last paragraph: this section would need sensitivity tests to better demonstrate that  $R^*$  does not represent a great uncertainty.

As answered previously, by carrying out sensitivity tests (Table R2.1), we find that 2x higher R\* leads to annual NH<sub>3</sub> emission decrease by approximately 30 %, and 2x lower R\* leads to 27 % higher emissions. The annual effect is smaller than the instantaneous response because lower emissions tend to be more sustained and vice versa.

P19 section 4.3.2: In this section a sensitivity to pH would be interesting to show to illustrate the possible effect of changing the manure pH by +-1 point.

We carry out a set of sensitivity tests (Table R2.1). We find that increasing pH from 8.5 to 9.5 causes annual NH<sub>3</sub> emission to increase by 5.8 %, while a decrease of pH to 7.5 leads to a decline of emission by 15.9 % (as described above). As with R\*, the sensitivity to pH is smaller for annual emissions as compared with instantaneous emission. More detailed discussion can be seen in the reply to Reviewer 1.

Table R2.1 Sensitivity test for model parameters for global application of the model.

Parameter	Value tested Value chan		ΔNH <sub>3</sub> emission %	
<sup>a, b</sup> Indoor resistance,	16700 s m <sup>-1</sup> (base)	1 x	0.0 %	
R*	8350 s m <sup>-1</sup>	0.5 x	<sup>a</sup> 27.1 %	a, b 8.5 %
K	33400 s m <sup>-1</sup>	2 x	a -30.6 %	a, b -6.4 %
	8.5 (base)	1 x	0.0	) %
a, b, c Manure pH (H <sup>+</sup> )	7.5	0.1 x	-15.9 %	
	9.5	10 x	5.8 %	
b, c Runoff coefficient, R <sub>runoff</sub>	1 % mm <sup>-1</sup> (base)	1 x	0.0 %	
	0.5 % mm <sup>-1</sup>	0.5 x	16.5 %	
	2 % mm <sup>-1</sup>	2 x	-11.8 %	
	11.2 Tg N year <sup>-1</sup> (base)	1 x	0.0 %	
a, b, c Excreted nitrogen	10.1 Tg N year-1	0.9 x	-12.3 %	
a D	12.3 Tg N year <sup>-1</sup>	1.1 x	12.6 %	

<sup>&</sup>lt;sup>a</sup> Parameters affect NH<sub>3</sub> emissions from housing. <sup>b</sup> Parameters affect NH<sub>3</sub> emissions from land spreading of chicken manure. <sup>c</sup> Parameters affect NH<sub>3</sub> emissions from backyard chicken.

## FIGURES AND TABLES

Fig 1: explain meaning of arrows

The arrows in Fig. 1 represent the nitrogen flows from chicken farming. We will update the figure caption of Fig. 1.

Fig 2: I would suggest adding flows in and out of the farm. In addition, an arrow for dilution through ventilation pointing towards INDOOR NH3 LEVELS may be considered. Watch out that the volatilisation flux is bi-directional. An arrow downwards should be shown.

Figure 2 shows critical processes of NH<sub>3</sub> emissions from chicken houses, which originates from chicken excretion. As we have not simulated other flows of N into our model out of the farm, we consider it better not to include such arrows. Process 1 represents the input of model that the nitrogen is in the form of UA from poultry excretion, and process 6 shows that the NH<sub>3</sub> emission is released from the houses to the outside atmosphere through ventilation (a flow out). The indoor NH<sub>3</sub> levels were simply calculated by dividing the NH<sub>3</sub> left in the house by the volume of the house. It may be noted that the arrow for process 6 is already connected to process 5.

Yes: we appreciate NH<sub>3</sub> fluxes can, in general, be both bi-directional, i.e. emission, or the reverse, deposition, and are dependent on the NH<sub>3</sub> concentrations in the surface source material and the overlying atmosphere. To reflect this point, we have referred at Eq. 7 to the study of Sutton et al. (2013) which considers this in detail. That paper also distinguishes between sources which are bi-directional (land surfaces) versus sources which are in effect only ever unidirectional (animal houses). For the situations in this study, i.e. NH<sub>3</sub> fluxes from N-rich animal excreta, we considered that chicken excretion is a strong source of NH<sub>3</sub> emissions from the surface, so we simplified the model to a uni-directional scheme. (We can envisage no practical case where outdoor atmospheric NH<sub>3</sub> concentrations would be larger than at the surface of chicken excreta). In order to be consistent with the model description, we do not include a downwards arrow in this situation.

Fig 3: It is unclear how the UA factors were calculated. 3a: could you give a hint on the significance of the difference between the two curves?

Figure 3a shows the relationship between the T factor of Elliot and Collins (1982) (red line) and that derived from the AFO experimental data (Section 2.2.1). (blue line). The blue line represents the least squares best-fit to the AFO data using a polynomial function of the form used by Elliot and Collins. It is possible to test whether the line of Elliot and Collins is significantly different from the data, by considering whether the mean difference (from the red line to points is significantly different from zero. For n=21, the mean difference in factor T between the red line and the data is 0.037 +/- 0.011 (standard error) which is significantly

different to zero with P>99% confidence. The value of Elliot and Collins is therefore significantly different from the AFO dataset.

Fig 4d and 5d: I would suggest showing also on the same graph the ammonia concentration at z0 (the compensation point) as it would give ground to better understand the NH3 emissions dynamics.

We will update the figures to include  $NH_3$  concentration at  $z_0$ .

Fig7: please explicit the fact that the curves are evaluated for yearly datasets. I suggest showing also total UAN remaining before cleaning to show any N-limiting effect on Pv. I also suggest rephrasing: 'NH3 volatilization rate Pv(%) for 4 different RH and Ta regimes....'

Agree. We will update Fig .7. We change "Curves that represent 4 different regimes from Fig. 6." into "Curves that represent  $NH_3$  volatilization rate  $P_V$  (%) for 4 different temperature and RH regimes based on annual steady-state simulations (see Fig. 6)."

Table 1: I would suggest adding percentage of N loss for each production system. In addition, you may consider getting rid of unneeded precision in emission numbers.

Agree. We will update manuscript Table 1.

Table R2.2 (manuscript Table 1) Excreted nitrogen from housed and backyard chicken, and estimated NH<sub>3</sub> emissions from each practice (global estimates for 2010). Uncertainty indicate the combined uncertainty ranges based on model sensitivity tests (Table R2.1).

Production system	Total excreted nitrogen (Tg N)	Practice	Total emission (Tg N)	Average P <sub>V</sub> (%)
Broiler and layer	9.0 [±0.9]	Housing	2.0 [±0.6]	22 [±7] %
		Land spreading	2.7 [±0.5]	39 [±7]* %

Backyard chicken	2.2 [±0.2]	Left on land	0.7 [±0.2]	32 [±7] %
Total	11.2 [±1.1]		5.5 [±1.2]	49 [±11] %

<sup>\*</sup> Based on the excreted N remaining (i.e., 7.0 Tg N) after NH<sub>3</sub> volatilization from housing.

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# A climate-dependent global model of ammonia emissions from chicken farming

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#### 10 Abstract

Ammonia (NH<sub>3</sub>) has significant impacts on the environment, which can influence climate and air quality, and cause acidification and eutrophication in terrestrial and aquatic ecosystems. Agricultural activities are the main sources of NH<sub>3</sub> emissions globally. Emissions of NH<sub>3</sub> from chicken farming are highly dependent on climate, affecting their environmental footprint and impact. In order to investigate the effects of meteorological factors and to quantify how climate change affect these emissions, a process-based model, AMmonia-CLIMate-Poultry (AMCLIM-Poultry) has been developed to simulate and predict temporal variations in NH<sub>3</sub> emissions from poultry excretion, here focusing on chicken farms and manure spreading. The model simulates the decomposition of uric acid to form total ammoniacal nitrogen which then partitions into gaseous NH<sub>3</sub> that is released to the atmosphere at hourly to daily resolution. Ammonia emissions are simulated by calculating nitrogen and moisture budgets within poultry excretion, including a dependence on environmental variables. By applying the model with global data for livestock, agricultural practice and meteorology, we calculate NH<sub>3</sub> emissions from chicken farming at global scale (0.5° resolution). Based on 2010 data, the AMCLIM-Poultry model estimates NH<sub>3</sub> emissions from global chicken farming of  $5.5 \pm 1.2$  Tg N yr<sup>-1</sup>, about 13 % of the agriculture-derived NH<sub>3</sub> emissions. Taking account of partial control of the ambient environment for housed chicken (layers and broilers), the fraction of excreted nitrogen emitted as NH<sub>3</sub> is found to be up to three times larger in humid tropical locations than in cold or dry locations. For spreading of manure to land, rain becomes a critical driver affecting emissions in addition to temperature, with the emission fraction being up to five times larger in the semi-dry tropics than in cold, wet climates. The results highlight the importance of incorporating climate effects into global NH<sub>3</sub> emissions inventories for agricultural sources. The model shows increased emissions under warm and wet conditions, indicating that climate change will tend to increase NH<sub>3</sub> emissions over the coming century.

### 1 Introduction

Ammonia (NH<sub>3</sub>) is the primary form of reactive nitrogen (N<sub>r</sub>) which has significant impacts on the environment (Galloway et al., 2003; Sutton et al., 2013). Following its emission to the atmosphere, NH<sub>3</sub> readily reacts with gas phase acids to form particulate ammonium aerosols and may also condense onto existing particles (Fowler et al., 2009; Hertel et al., 2011). Gaseous NH<sub>3</sub> reacts with sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) and nitric acid (HNO<sub>3</sub>), which leads to formation of ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) aerosols, respectively (Pinder et al., 2007, 2008; Hertel et al., 2011). These particles influence the radiation balance of the Earth by scattering light and altering the Earth's reflectivity (Xu and Penner, 2012), and also adversely affect regional air quality and human health (Brunekreef and Holgate, 2002; Pinder et al., 2007, 2008). The lifetime of atmospheric NH<sub>3</sub> is relatively short (hours to days) as it is removed rapidly by dry and wet deposition, or converted

to ammonium aerosols (Hendriks et al., 2016). Consequently, it is usually removed close to its source. In terrestrial ecosystems, acute exposure to NH<sub>3</sub> can cause visible foliar injury, reducing vegetation's tolerance to pests and diseases, especially for native plants and forests (Krupa 2003; Stulen et al., 1998; Sutton et al., 2011). Once deposited in water, NH<sub>3</sub> can result in acidification and eutrophication (Sutton et al., 2011). Excess N<sub>r</sub> input causes algal blooms in vulnerable aquatic ecosystems, which harms local biodiversity.

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The dominant source of NH<sub>3</sub> emission is from agricultural activities including animal housing, manure storage, and fertiliser usage for arable lands and crops. In western countries, approximately 80-90 % of atmospheric releases are from agriculture (Sutton et al., 2000; Hertel et al., 2011); a major source of NH<sub>3</sub> emission is from livestock waste. Oenema et al. (2007) estimated that NH<sub>3</sub> emissions cause a loss of approximately 19 % of nitrogen from livestock housing and manure storage, with a further 19 % being lost following the land application of manure. Previous studies that quantified NH<sub>3</sub> emissions from livestock have made estimations mainly by empirical methods. Emission factors were used, assuming fixed values for nitrogen volatilization rates, varying by animal type and management practices. For example, Misselbrook et al. (2000) derived NH<sub>3</sub> emission factors for major animals under various farming practices in UK agriculture. The advantage of this method is the relative simplicity for calculations. However, these emission factors only include climatic effects to a small extent. Using a fixed number to describe the fraction of excreted nitrogen that volatilises as NH<sub>3</sub> does not always provide a realistic value under all environmental conditions and may cause large uncertainties in large scale estimations (e.g., when considering global scale estimates). Sommer and Hutchings (2001) reviewed a range of empirical models that were produced to predict NH<sub>3</sub> volatilization from slurry application to land. These models have experiment-derived equations. However, only one or two factors the effect of temperature and slurry dry matter content were studied and the interactions between these parameters were not investigated.

Another method for estimating NH<sub>3</sub> emission from livestock is to use process-based models based on a theoretical understanding of relevant processes, building on foundations developed for field sources (Sutton et al., 1995b; Nemitz et al., 2001; Móring et al., 2016). Pinder at al. (2004) developed a process-based model for simulating NH<sub>3</sub> emissions from dairy cows, and the modelled NH<sub>3</sub> volatilization fraction from grazing, manure spreading and storage was shown to be reasonable compared to independent experimental data. Previous process modelling efforts for bird sources have focused on native seabird populations (Riddick et al., 2016, 2018), using these as a natural laboratory to study the effect of global climate differences on NH<sub>3</sub> emissions, supported by a programme of measurements through different climates (Blackall et al., 2007; Riddick et al. 20152012). Process-based models consider the effects of meteorological variation on the formation of NH<sub>3</sub> from an N<sub>r</sub> source, allowing calculation of NH<sub>3</sub> emissions that vary temporally and spatially. They can be extended to investigate the influences of various environmental conditions. However, as more complicated parameterizations are included in process-based models, more detailed inputs are required, and lack of input data may limit the model's ability to obtain better results.

Ammonia emissions from animal waste are understood to be highly climate-sensitive. For example, Sutton et al. (2013) showed a factor of nine increase in emission rates between 5 °C and 25 °C, with additional effects from humidity and precipitation (Riddick et al., 2017). Poultry numbers have increased roughly five-fold over the last 50 years (FAO, 2018), with chicken being the largest fraction. Global usage of poultry manure for land spreading increased from an estimated 5.0 Tg N yr<sup>-1</sup> in 2000 to 6.3 Tg N yr<sup>-1</sup> in 2010 (FAO, 2018). However, limited research has attempted to determine the magnitude of global NH<sub>3</sub> emissions from chicken farming whilst also considering climatic effects. In this study, a process-based model, AMmonia-CLIMate-Poultry (AMCLIM-Poultry) has been developed to simulate and predict temporal variations in NH<sub>3</sub> emissions from

three major chicken production systems: (a) broilers, (b) layers and (c) backyard chicken, focusing on chicken housing and land spreading of manure. The overarching goals of this study are to develop a process-based model and to apply it at global scale, to produce improved NH<sub>3</sub> emission estimates under influences of various meteorological factors, and to estimate total NH<sub>3</sub> emissions and their distribution for the present-day (year 2010) for chicken farming globally. Future work will quantify the estimated response of NH<sub>3</sub> emissions to climate change, the potential for year-to-year variability, and the implications for NH<sub>3</sub> emissions from other livestock sectors.

### 2 Methods and Materials

### 2.1 Model description

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Figure 1 shows <u>agricultural</u> activities <u>related to in which</u> chicken litter <u>as-is</u> a source of NH<sub>3</sub> emission in <u>agricultural practices</u>.

Nitrogenous manure can be used as fertilisers on land or be stored for future <u>usageuse</u>. Typically, litter collected from chicken houses is spread on <u>soils for fertilising cropsarable lands</u> at the start of planting period, while excretion from backyard systems are applied fresh to fields or left on pastures and other ground. Ammonia can be released to the atmosphere through each of these activities. In this study, we developed the process-based AMCLIM-Poultry model to quantify NH<sub>3</sub> emissions from chicken farming, focusing on housing and manure land spreading. For this purpose, it is assumed in the model that emissions from stored manure occur within the animal house ('in-house storage') or do not behave significantly differently. The <u>uncertainties associated with this simplification are considered in Sect. 4.3.1.</u>

The model has been developed from the GUANO model (Riddick et al., 2017) that , which simulatesed NH<sub>3</sub> emissions from wild seabird colonies, which provides a starting point for AMCLIM-Poultry. Both models simulate  $N_r$  through the decomposition processes that uric acid (UA, solid/aqueous phase) in excreta hydrolyses to form total ammoniacal nitrogen (TAN = NH<sub>3</sub> + NH<sub>4</sub><sup>+</sup>, aqueous phase), which then partitions to form gaseous NH<sub>3</sub> that is released to the atmosphere (Fig. 2). Major advances in the present study using AMCLIM-Poultry compared with the GUANO model include:

- a) There is a he distinction between indoor and outdoor simulations, which represent different practices and production systems under different environmental conditions (housing birds, manure spreading, backyard birds). emissions,
- <u>b)</u> <u>conservation Flow of nitrogen is conserved between these the different stages of housing and manure spreading following excretion, which reflects the reality that nitrogen emitted as NH<sub>3</sub> cannot be emitted again.<sub>5</sub></u>
- c) a-A new approach is developed to simulate indoor emissions. Environmental conditions of houses and a new parameterization for UA hydrolysis are generalised from measurement datasets. Ammonia volatilized from the animal waste at the surface is determined by a parameterized resistance term that is derived from measurements.
- <u>d)</u> <u>and the Land linking of land</u> spreading of chicken manure <u>is linked</u> to the timing of agricultural cropping cycles, <u>which</u> <u>allows a better estimate of NH<sub>3</sub> emissions and its temporal variations.</u>

We used chicken exerction exerctal derived nitrogen as an input (described in Sect. 2.4.1), and incorporated meteorological factors to predict temporal variations of the NH<sub>3</sub> emissions. The simulations followed N, through the decomposition processes that uric acid (UA, solid/aqueous phase) in excretion hydrolyses to form total ammoniacal nitrogen (TAN = NH<sub>3</sub> + NH<sub>4</sub>; aqueous phase), which then partitions to form gaseous NH<sub>3</sub> that is released to the atmosphere (Fig. 2). The quantitative equations used in the model are described below using SI units (except for mass unit, for which gram was used instead of

kilogram). In the simulations, <u>T</u>the model was operated with an hourly time step for outdoor <u>emissions simulations</u> and a daily time step for indoor <u>emissions simulations with corresponding units being converted</u>.

### 2.1.1 Mass balance of nitrogen components

The AMCLIM-Poultry model simulates masses for N-containing components (UA, TAN) within the chicken farming system (chicken houses; backyard chickens; and chicken manure spreading), and flows between these pools (Fig. 1). The mass per unit area of excretion ( $M_{excretion}$ , g m<sup>-2</sup>; all model variables are described, with units, in the Appendix) over time-step  $\Delta t$ , is calculated following Eq. (1):

$$M_{excretion}(t + \Delta t) = M_{excretion}(t) + \frac{F_e}{f_N} \Delta t,$$
 (1)

where  $F_e$  (all nitrogen flows have units of g N m<sup>-2</sup> s<sup>-1</sup>) is total nitrogen excretion rate from chicken and  $f_N$  (g N g excretion<sup>-1</sup>) is the nitrogen content of excretion. The evolution of UA mass ( $M_{UA}$ ; all nitrogen pool masses have units of g N m<sup>-2</sup>) over timestep  $\Delta t$ , is calculated following Eq. (2):

$$M_{UA}(t + \Delta t) = M_{UA}(t) + (F_e f_{UA} - F_{TAN}) \Delta t, \tag{2}$$

where  $f_{UA}$  is the UA fraction in the excretion, and  $F_{TAN}$  is the <u>fluxconversion rate</u> of TAN that is decomposed from UA hydrolysis of UA to TAN.

Similarly, the mass of TAN  $(M_{TAN})$  is calculated following Eq. (3):

$$M_{TAN}(t + \Delta t) = M_{TAN}(t) + (F_{TAN} - F_{NH_3})\Delta t, \tag{3}$$

where  $F_{\text{NH3}}$  is the net rate of conversion of TAN to gaseous NH<sub>3</sub> that is emitted to the atmosphere. All pools are set to zero when there is an emptying event for housing.

### 2.1.2 Process-based simulation of nitrogen pathways

For each emission context (i.e., animal housing, backyard birds, manure spreading), the AMCLIM-Poultry model includes three key steps: conversion of UA to TAN, equilibrium between aqueous phase TAN and gaseous NH<sub>3</sub> in the litter, and volatilization of NH<sub>3</sub> from the litter surface to the atmosphere (Fig. 2). Uric acid is converted The hydrolysis of UA to TAN by hydrolysis, which is strongly affected by temperature, the pH of the substrate, and the relative humidity (RH) of the chicken house atmosphere (Elliott and Collins, 1982; Elzing and Monteny, 1997; Koerkamp, 1994). The production rate of TAN is determined from the UA mass and the conversion rate (K), which is a function of these three factors:

$$F_{TAN} = M_{UA}K_{(T,pH,RH)} \tag{4}$$

The maximum estimated production rate is 20 % per day at 35 °C, pH 9.0, and RH 80 % (Elliot and Collins, 1982). The combined influence of these three factors is the product of a series of conversion rate functions:

$$K_{(T,pH,RH)} = 0.2 k_{pH} k_T k_{RH}$$
 (5)

Gas phase NH<sub>3</sub>, held within the litter pore spaces, is in equilibrium with TAN that depends upon the litter pH and temperature response of combined Henry and disassociation equilibria (Eq.( $\frac{56}{9}$ )) (Nemitz et al., 2000). The gas phase concentration of NH<sub>3</sub> in air ( $\chi$ ) at the surface is proportional to the aqueous phase ratio  $\Gamma = [NH_4^+]/[H^+]$  of the chicken litter, which is calculated from Eq. ( $\frac{56}{9}$ ) and Eq. ( $\frac{67}{9}$ ):

$$\chi = \frac{161500}{T} \exp\left(\frac{-10378}{T}\right) \Gamma, \tag{56}$$

$$\Gamma = \frac{[NH_4^+]}{[H^+]} \frac{[NH_4^+]}{[H^+]} = \frac{[TAN]}{K_{NH_4^+} + [H^+]} = \frac{M_{TAN}}{V_{H_2O}(K_{NH_4^+} + [H^+])} \frac{M_{TAN}}{V_{H_2O}}, \tag{67}$$

where [NH<sub>4</sub>+] is in units of g N ml<sup>+</sup> and  $V_{H_2O}$  (ml m<sup>-2</sup>) is the volume of water in the litter, and  $K_{NH_4+}$  is the dissociation constant of NH<sub>4</sub>±. Ammonia volatilises to the atmosphere from the surface at a rate ( $F_{NH_3}$ ) that can be determined by assuming a resistance type model: using gas concentrations at two vertical levels constrained by a set of resistances (Sutton et al., 2013), which is calculated from Eq. (78):

$$F_{NH_3} = \frac{[\chi(z_{o'}) - \chi(z)]}{[R_a(z) + R_b]}, \tag{78}$$

where  $\chi(z_{o'})$  represents the concentration at the surface, and  $\chi(z)$  represents the concentration at a reference height. Equation (7) is the general formula. For in-house application of the model,  $\chi(z)$  is taken as representative of well mixed indoor concentration of NH<sub>3</sub> in chicken house. For outdoor application of the model, the reference height is taken 10 m above ground.  $R_a$  and  $R_b$  are the aerodynamic and boundary layer resistances, respectively. This broad resistance approach is applicable for manure spread in the field and is also applied for backyard birds. For emissions from housed chicken resistance in chicken houses, a modified approach is needed as described in Sect. 2.2.2.

### 2.2 Simulations for chicken housing

Figure 2 illustrates the process pathways through which NH<sub>3</sub> volatilises from the N-rich chicken excretion to the exterior atmosphere. We assumed 60 % of excreted nitrogen is in the form of UA (*f<sub>UA</sub>* = 0.6), which accounts for approximately 3-8 % of the chicken excretion (Nahm, 2003). The remaining 40 % of excreted nitrogen is <u>assumed to be in all</u> other forms that <u>are not liabledo not lead</u> to significant NH<sub>3</sub> emissions. Uric acid accumulates in the litter of the chicken house until it converts to TAN by bacterial ammonification, with TAN concentrations in equilibrium with the litter pore space concentration of gaseous NH<sub>3</sub>. Ammonia is then emitted from the surface, which builds up the indoor NH<sub>3</sub> levels within the house through mixing. Meanwhile, <u>as NH<sub>3</sub> is removed continuously through ventilation because</u> the indoor NH<sub>3</sub> concentration must be controlled below a certain level, <u>ventilation continuously removes NH<sub>3</sub> and brings fresh air which dilutes the NH<sub>3</sub> concentrations.</u>

We used the monitored data from Animal Feeding Operations (AFOs, 2012) to simulate site-specific NH<sub>3</sub> emissions from chicken houses. The data were gathered by the US Environmental Protection Agency (EPA) as a study of emissions from different types of livestock from 2007-2010 (Cortus et al., 2010; Jin-Qin Ni et al., 2010; Wang et al., 2010). As shown in Table S1 (Supplementary Sect. 1), two broiler houses and four layer houses from three US farms at different sites were selected for this study. We used daily mean animal data, environmental data, and indoor NH<sub>3</sub> concentrations (measured at 2 - 2.5 m above the ground, representative of well mixed air in the chicken house) from these sites. Animal data included bird numbers, body weight, and biomaterial data for each house; and environmental data included temperature, relative humidity

for natural (outdoor) and indoor conditions, and the interior ventilation given as an airflow rate in m<sup>3</sup> s<sup>-1</sup>. We filled up missing environmental data to keep simulations continuous by using a linear interpolation method when measurements were unavailable to keep simulations continuous. Excreted nitrogen was determined from the animal data and was used as an input to the model, together with the indoor environmental data. As the AMCLIM-Poultry model does not simulate evaporation from litter in houses, we determined th Thee excretion water content ( $M_{H_2O}(e)$ , g m<sup>-2</sup>) that determines the TAN concentration of litter is dependent based on the equilibrium moisture content ( $m_{E_1}$  %) of the litter, which is calculated from Eq. (89):

$$M_{H_2O}(e) = \frac{m_E}{100} \cdot M_{excretion},$$
 (89)

where  $m_E$  is calculated following the Eq. (910):

$$m_E = \left[ \frac{-\ln\left(1 - \frac{RH}{100}\right)}{0.0000534 \times T} \right]^{\frac{1}{1.41}},\tag{910}$$

where *RH* (%) is the relative humidity, and *T* (K) is the temperature (Elliott and Collins, 1982). Equation 10 is based on the hygroscopicity of chicken litter and accounts for the moisture absorbed by the litter as it reaches an equilirium state, which is dependent on temperature and RH.

### 2.2.1 Parametrization of UA hydrolysis rate for chicken housing

The hydrolysis of UA to TAN plays a crucial role in affecting NH<sub>3</sub> emissions. The rate of conversion of UA to TAN is often the rate-limiting process that determines the overall rate of conversion of nitrogen excreted by chicken into NH<sub>3</sub> emissions. The parametrization of UA to TAN conversion is therefore very important for the overall model performance.

In the study of Elliott and Collins (1982), a chicken litter model was used to investigate the UA hydrolysis rate. They set the base level conversion rate to 20 % over a 24-hour period under optimal conditions (pH = 9,  $T \ge 35$  °C, RH  $\ge 80$  %), then produced empirical functions to account for the influence of these three factors. In order to evaluate the validity of these empirical functions, specifically temperature and RH effects, we analysed the AFO measurements for two layer houses from the US EPA dataset (Table S1), starting from the date that litter was cleaned out from the houses. We assumed an equilibrium state between the production of TAN and NH<sub>3</sub> emission, and a constant litter pH of 8.5. It should be noted that the equilibrium state does not always apply, but it is a useful assumption for parameterization, and the introduced uncertainty is discussed in Sect. 4.1.21. The temperature dependence was derived from measurements when RH was over 80 %, and the RH dependence was derived from measurements that were normalised by the temperature dependence. We used these data to update the empirical functions of Elliott and Collins (1982) that parameterize the UA hydrolysis rate (see Sect. 3.1.2).

The temperature and RH dependence of UA hydrolysis rate derived from using the AFO monitored data are shown in Fig. 3, where they are compared to functions from Elliott and Collins (1982). The new temperature dependence follows an exponential relationship, and is normalised to the maximum rate at 35 °C:

$$30 k_T = \frac{exp^{(0.149(T-273.15)+0.49)}}{exp^{(0.149(35)+0.49)}} (11)$$

The new RH dependence increases linearly as RH increases, reaching the maximum rate of 1 at RH 80 %:

$$k_{RH} = \begin{cases} 0.0125RH - 0.0014, & if \ 0 < RH < 80\% \\ 1, & if \ 80\% \le RH \end{cases}$$
 (12)

Within the range of RH 0~40 %, the function is extrapolated due to the limited data at these conditions (Fig. 3b). The new RH dependence is parameterized directly as a function of RH rather than the excretion moisture content because it is envisaged that fresh excretion reaches an equilibrium moisture within a few hours, and it is a representative simplification to use the RH data as the model is run on a daily time-step.

We used the pH dependence for the range of 5.5 to 9.0 from the Elliott and Collins (1982) study:

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$$k_{pH} = \frac{1.34(pH) - 7.2}{1.34(9) - 7.2} \tag{13}$$

A fixed pH of 8.5 that is the typical value of poultry manure (Elliott and Collins, 1982; Sommer and Hutchings, 2001) was used for the simulations. We did not include a dynamical scheme for determining pH influenced by the UA hydrolysis (cf. Móring et al., 2016), which is a practicable simplification for a global model.

#### 2.2.2 Inversion of resistance within chicken houses to develop R\* parametrization of chicken houses

The NH<sub>3</sub> flux from an unvegetated surface to the atmosphere is mainly constrained by two terms: aerodynamic resistance ( $R_a$ ) and boundary layer resistance ( $R_b$ ) (Wesely, 1989). Outdoors, both these resistances are related to meteorological conditions and can be calculated. However, values of  $R_a$  and  $R_b$  within chicken houses remain unknown due to the lack knowledge of turbulence for indoor conditions. We estimated the overall indoor resistance, termed  $R^*$ , which includes  $R_a$ ,  $R_b$  and also the resistance of litter, by inversion of the measured AFO data. As shown by steps 4, 5 and 6 in Fig. 2, the interior NH<sub>3</sub> level within a chicken house is determined by the source flux from the litter surface and the removal flux through ventilation. Mathematically, the total flux of NH<sub>3</sub> ( $F_{Surface}$ , g N s<sup>-1</sup>) from the surface is expressed as Eq. (1014):

$$F_{surface} = \left(\frac{\chi_{surface} - \chi_{in}}{R^*}\right) \cdot S, \tag{1014}$$

where  $\chi_{surface}$  (g m<sup>-3</sup>) is the in-house value of  $\chi(z_{o'})$ , i.e, the gaseous NH<sub>3</sub> concentration at the litter surface and  $\chi_{in}$  (g m<sup>-3</sup>) is the indoor NH<sub>3</sub> concentration of the house assuming a complete mixing of air inside the chicken house.  $R^*$  (s m<sup>-1</sup>) is the indoor resistance, and S (m<sup>2</sup>) is the surface area of the house. The NH<sub>3</sub> removal ( $F_{removal}$ , g N s<sup>-1</sup>) through ventilation is expressed as Eq. (4+15):

$$F_{removal} = Q\left(\chi_{in} - \chi_{out}\right),\tag{1115}$$

where  $\chi_{out}$  (g m<sup>-3</sup>) is the free-atmosphere NH<sub>3</sub> concentration.  $\chi_{out}$  is set to be 0.3 µg m<sup>-3</sup>, which is normally much lower than the indoor concentration. Q (m<sup>3</sup> s<sup>-1</sup>) represents the ventilation rate. Therefore, by mass conservation, we can relate indoor NH<sub>3</sub> concentrations and the interior air volume V (m<sup>3</sup>), to surface emissions and losses through ventilation:

$$V \frac{d\chi_{in}}{dt} = F_{surface} - F_{removal}$$

$$= \left(\frac{\chi_{surface} - \chi_{in}}{R^*}\right) \cdot S - Q\left(\chi_{in} - \chi_{out}\right)$$
(1216)

For inversion of  $R^*$ , we used the data for two layer houses at NC2B, which had clearly reported house emptying dates and had fewer missing measurement data. The simulation period started from the day when litter was cleaned out, and each nitrogen pools was re-initialised. For the inversion, we we assumed the house reached steady-state (hence the LHS of eq. (12) is zero) after a period of simulation for three days, and the term  $Q\chi_{out}$  has been neglected due to its small magnitude. Subsequently, the resistance can be calculated from Eq. (1317):

$$R^* = \frac{(\chi_{surface} - \chi_{in}) \cdot S}{Q\chi_{in}}$$
 (1317)

To develop this parametrization, the gas phase NH<sub>3</sub> concentration at the surface ( $\chi_{surface}$ ) was simulated by the AMCLIM-Poultry model and the NH<sub>3</sub> concentration within the house and ventilation were taken from the AFOs monitored data.

#### 2.3 Simulations of NH<sub>3</sub> emission from chicken manure spreading

10 Simulations for spreading of chicken manure to fields followed the processes of nitrogen pathways which are similar to the housing simulations. Nevertheless, there are several key points need to be clarified. Firstly, cContrary to the housing, the simulation of NH<sub>2</sub>-emissions from the spreading of chicken manure to fields is different due to the following points. First, the amount of water in the system  $(M_{HaQ}, g m^2)$  is is calculated in a different way related to the environmental conditions, which includes rainfall, evaporation and runoff, rather than to only depend on litter moisture. Secondly, runoff takes place during rain events and is a major loss of nitrogen. Thirdly, aerodynamic resistance  $(R_a)$  and boundary layer resistance  $(R_b)$  that 15 determines the magnitude of NH<sub>3</sub> emissions are directly calculated from meteorological variables instead of being parameterized (Nemitz et al., 2001; Seinfeld and Pandis, 2016; Riddick et al., 2017). Details are given in Supplementary Sect. 2. Fourthly, we only simulate processes taking place in manure and do not simulate interactions with soils. We consider it reasonable as chicken manure is mainly applied on the land surface because it is dry and not physically mixed with underlying 20 soils based on the assumption of a simple application scenario. In addition, simulating soil processes would require a much more detailed characterization of soil chemistry, which might only be achieved by using sophisticated land models that are beyond the scope of this study.

The amount of water in the litter ( $M_{H_2O}$ , g m<sup>-2</sup>) is calculated from: related to the outdoor environment (i.e. precipitation, evaporation and runoff):

$$25 \quad M_{H_{2}O}(t + \Delta t) = \begin{cases} M_{H_{2}O}(t) - M_{available \, water} + \left( F_{H_{2}O}(rain) - F_{H_{2}O}(evap) \right) \Delta t + M_{H_{2}O}(e), if \\ M_{H_{2}O}(t) - M_{available \, water} + \left( F_{H_{2}O}(rain) - F_{H_{2}O}(evap) \right) \Delta t > 0 \\ M_{H_{2}O}(e), if \\ M_{H_{2}O}(t) - M_{available \, water} + \left( F_{H_{2}O}(rain) - F_{H_{2}O}(evap) \right) \Delta t \leq 0 \end{cases}$$

$$M_{available \, water} + \left( F_{H_{2}O}(rain) - F_{H_{2}O}(evap) \right) \Delta t, \qquad (1418)$$

where  $F_{H_2O}(rain)$  (g m<sup>-2</sup> s<sup>-1</sup>) and  $F_{H_2O}(evap)$  (g m<sup>-2</sup> s<sup>-1</sup>) are the precipitation rainfall and evaporation, respectively, and  $M_{available\ water}$  (g m<sup>-2</sup> s<sup>-1</sup>) is the water available for run-off. It should be noted that the amount of water of the system in the manure should not be less than the excretion water content, which is the equilibrium moisture content dependent on environmental conditions. R-The maximum amount of water that can be absorbed by the manure, which was assumed to be a factor of 2 × of the mass of excretion (Riddick et al., 2017). The water left in the system is the amount of water available for runoff ( $M_{available\ water}$ , g m<sup>-2</sup>):

(15)

Second, runoff takes place under natural conditions especially during rain events and is a major loss of nitrogen.

In the model, the immediate runoff ( $M_{N-runoff}$ , g m<sup>-2</sup>) is derived from the a runoff coefficient multiplied by the nitrogen pools:

$$M_{N-runoff} = R_{runoff} \cdot M_{N}, \tag{1619}$$

where the  $M_N$  (g m<sup>-2</sup>) is the amount of each N-containing components, and  $R_{runoff}$  is the runoff coefficient that is a function of the amount of water within the nitrogen pools available for runoff ( $Q_{available\ water}$ , mm):

$$R_{runoff} = Q_{available \, water} \cdot r_{N}, \tag{1720}$$

where  $r_N$  (mm<sup>-1</sup>) represents the wash off factor, and constant values was used of 1 and 0.5 % mm<sup>-1</sup> for nitrogen and manure, respectively (Riddick et al., 2017). The maximum amount of water that can be absorbed by the manure, which was assumed to be a factor of 2 × of the mass of exerction (Riddick et al., 2017). The water left in the system is the amount of water available for runoff ( $M_{available water}$ , g m<sup>-2</sup>) is determined by subtracting the water absorbed by the manure from rainfall:

$$M_{available \, water} = F_{H_2O}(rain)\Delta t - 2 \times M_{excretion}$$
 (1521)

The maximum amount of water that can be absorbed by the manure was assumed to be 2x of the mass of excretion (Riddick et al., 2017).

Third, the resistances including aerodynamic ( $R_a$ ) and boundary layer resistance ( $R_b$ ) were directly calculated from meteorological variables instead of being parameterized (Nemitz et al., 2001; Seinfeld and Pandis, 2016; Riddick et al., 2017).

#### 2.4 Global applications

#### 20 **2.4.1 Model input**

In order to quantify the NH<sub>3</sub> emission from global chicken farming, We we applied the AMCLIM-Poultry model at the global scale to quantify the NH<sub>3</sub> emissions from global chicken farming. The model used the FAO (Food and Agricultural Organization of United Nations) global chicken density data and chicken excretion nitrogen data as input and was driven by the ECWMF ERA5 hourly meteorological data (ERA5, 2018). The model was run under a resolution of  $0.5^{\circ} \times 0.5^{\circ}$ , with the global chicken density data and nitrogen data being regridded to fit the  $0.5^{\circ}$  resolution.

The global population of chickens was based on FAOSTAT data for 2010 (FAOSTAT). The geographic distribution was based on the Gridded Livestock of the World (GLW) model, which produced density maps for the main livestock species based on observed densities and explanatory variables such as climatic data, land cover and demographic parameters (Robinson et al.,

2014). The chicken data were categorised into three production systems: broilers, layers and backyard chicken. Broilers and layers are major chicken types that are reared intensively in buildings and managed by farmers or livestock companies. The environment for rearing backyard chicken is varied and the density is lower compared with broilers or layers. The distinction in the global distribution of backyard and intensive systems was based on Gilbert et al. (2015). Birds in the intensive systems were further subdivided into broilers and layers using the procedure developed for the Global Livestock Environmental Assessment Model (GLEAM FAO, 2018). The GLEAM approach was also used to produce the nitrogen excretion maps, which were calculated as the difference between nitrogen intake and retention. The total nitrogen intake depends on feed intake and nitrogen content of the feed, while the retention is the amount of nitrogen that is retained in birds' tissues, either as live weight gain or production of eggs (FAO, 2018).

#### 2.4.2 Global upscaling for chicken housing

In chicken farms, the inside conditions can be distinct from the natural environment. The 'lower critical temperature' for chicken (i.e., the minimum managed temperature for optimum chicken performance) is approximately 16-20 °C (Gyldenkærne et al., 2005) which is much higher than of other livestock, such as cattle and sheep. Intensively managed chicken are typically kept in insulated buildings with forced ventilation and heating systems to help maintain fixed temperature throughout the year as far as feasible (Seedorf et al., 1998). To keep the ambient temperature within a recommended range, the house may be heated or ventilated in relation to outdoor temperatures. Heating occurs on cold days when temperature is low but not in other periods. Ventilation is to maintain a healthy condition for chicken's growth, and a minimum level is required, but also the ventilation should be below a certain rate to avoid induced draft in the house (Gyldenkærne et al., 2005).

For the modelling, the broilers and layers were assumed to be kept in buildings with adequate heating and ventilation systems. The density for broilers and layers was assumed to be 15 birds/m<sup>2</sup> and 30 birds/m<sup>2</sup>, respectively (Cortus et al., 2010; Jin-Qin Ni et al., 2010; Krause and Schrader, 2019; Wang et al., 2010). In the AMCLIM-Poultry model, tThe environmental parameters incorporated in the model are empirically derived from the indoor environment of chicken farms reported in the EPA dataset. The housing temperature is determined by the generalised relationships between indoor and outdoor/natural temperature shown in Fig. S1 (Supplementary Sect. 3), while the RH in the house is set to be identical to ambient RH as no obvious relationship 25 was found according to the EPA dataset. It is assumed that the temperature and ventilation rates of chicken houses are maintained as close as possible to a stable level throughout the day and are driven by the natural climatic conditions under local practice. There is no precipitable water in the house, so the water budget pool excludes precipitation and is purely related to the determined by excretion moisture. The litter in chicken houses was assumed to be removed once a year. The housing part simulation of the AMCLIM-Poultry model was operated at a daily time-step based atfor 2010, as the indoor conditions are derived from daily measurements. To calculate the varying impacts of emptying the chicken houses at different times of the year, we ran 12 different year-long simulations, each starting from a different month, i.e. from January to December, and assuming the chicken house had just been emptied. 12 simulations were run by assuming that chicken houses were emptied in different months for each simulation, i.e. from January to December, and the simulations started in corresponding month. The results were averaged and reported in this study.

# 2.4.3 Global upscaling for chicken manure spreading

As shown in Fig. 1, the manure from chicken farms are collected for applications spreading to fields, leading to NH<sub>3</sub> emissions. Typically, fertilising crops use manure from local farms. Therefore, we assumed the amount of nitrogen from chicken manure is only spread locally, and the simulations for each grid-cell are independent to the adjacent ones in terms of model input. This

assumption is considered to be valid at  $0.5^{\circ} \times 0.5^{\circ}$  resolution of the global model application (equivalent to 39 km  $\times$  55 km at  $45^{\circ}$  latitude), though cannot be automatically assumed when modelling at finer scales. The available nitrogen budgets were determined from the amount of nitrogen left, ensuring mass-consistency to account for NH<sub>3</sub> emitted in the housing simulations.

It should be emphasized that the global distribution of available nitrogen for land spreading of chicken manure may not completely coincide with global distribution of croplands or the global usage of inorganic nitrogen fertilisers. It is assumed in the model application here that chicken manure is only used must only take place in regions that have on arable lands, and the amount of nitrogen applied on land should not exceed the total manure-N application rates so there should not be any manure applications from intensively managed housed chicken regions with no farming practice. Meanwhile, there are thresholds for nitrogen applications for crops. If nitrogen application rates required to use the chicken manure on agricultural land exceed the maximum guided amount, it will have harmful or lethal effects on crops. Therefore, simply using the total available nitrogen from livestock manure as inputs to the land spreading part of the AMCLIM-Poultry model could cause error and not reflect reality.

To address these considerations, we defined the amount of nitrogen applied to crops as contributed nitrogen input. To estimate the contributed nitrogen input from chicken manure, we compared the available amount of chicken manure-N (nitrogen left in manure after being lost as NH<sub>3</sub> at housing period) to the total amount of manure-N for crops to identify places that would use chicken manure as fertiliser. Data of the total amount of manure-N used for crops and fertilising areas were used taken from West et al (2014). We chose six major crops for which chicken manure is ideal fertiliser, including barley, maize, potato, rice, sugar beet and wheat. We assumed the chicken manure is primarily applied to these six crops. For areas where available chicken manure-N does not exceed the total manure-N application, we calculated the contributed nitrogen input for individual crops by Eq. (1822):

$$N_{Crop\_Poultry} = N_{Soil\_PoultryAvailable} \cdot \frac{N_{Crop}}{N_{Total\_Manure}} \cdot \frac{1822}{N_{Total\_Manure}}$$

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Conversely, for areas where available nitrogen input from chicken exceeds the total manure-N application, the contributed nitrogen input is calculated from Eq. (1923):

$$25 \quad N_{Crop\_Poultry} = N_{Crop}, \tag{1923}$$

where  $N_{Crop\_Poultry}$  (g N m<sup>-2</sup>) is the amount of chicken manure-N application for individual crops,  $N_{Soil\_Poultr\_Availabley}$  (g N m<sup>-2</sup>) is the amount of available chicken manure-N,  $N_{Crop}$  (g N m<sup>-2</sup>) is the amount of total nitrogen application for individual crops,  $N_{Total\_Manure}$  (g N m<sup>-2</sup>) is the amount of total nitrogen application from manure for all crops. The excess nitrogen in these areas was considered to be applied to other crops. In regions where annual nitrogen applications are zero, we assumed the available chicken manure-N are untreated and left on land.

Planting and harvesting dates for crops are important parameters in the model because they determine the meteorological conditions of the crop growing period, which affects the temporal variations of NH<sub>3</sub> emission from land spreading. Fertiliser applied to land or crops is dependent on the timing of agricultural activities rather than being spread frequently. As a result, the NH<sub>3</sub> emission from fertiliser spreading usually shows strong seasonal variations due to the local farming practice. In this study, the The AMCLIM-Poultry model incorporates the planting and harvesting dates from the Crop Calendar Dataset for the

six major crops to make estimates (Sacks et al., 2010). We developed a relatively simple scenario for fertiliser manure applications that the chicken manure fertiliser was applied at the start of planting period. Timing of agricultural practices in the southern hemisphere is different from the northern hemisphere. The planting activities usually start in November or December, which causes that partial NH<sub>3</sub> emissions in these regions would occur in the next year. Similarly, manure spreading that took place in the last year can also result in emissions in the current year. Therefore, we ran the model for more than one year to keep an annual cycle of simulation period for each grid. It should be emphasized that our model scenario assumes a standard reference that all chicken manure is broadcast on the surface of bare agricultural fields, at the start of the cropping cycle. Other future scenarios could consider the effectiveness of management practices to mitigate NH<sub>3</sub> emission from the spreading of chicken manure (see Sect. 4.45).

As introduced in Sect. 2.4.1, backyard chicken is one of the major production systems included in the FAO chicken density dataset. In comparison with broilers and layers, backyard chicken is reared in residential lots rather than in insulated houses. According to the FAO statistics, there are two general ways of dealing with excretion from backyard chicken: daily spreading and leaving it on pastures. Consequently, the simulations for NH<sub>3</sub> emissions from backyard chicken were set to be under natural environments. Data for excreted nitrogen from backyard chicken from the FAO dataset were used as the nitrogen input to the model. The density was assumed to be 4 birds/m<sup>2</sup>. The meteorological inputs were the same as used in the simulations for chicken manure spreading for crops. The model was operated at an hourly time-step for a period of one year as an initialisation. The second-year simulation was for the study period of 2010.

#### 3 Results

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#### 3.1 Site simulations for chicken housing

## 20 3.1.1 Temperature of chicken houses

A generalised representation of indoor temperatures of chicken housing was empirically derived from the AFOs measurements from the three farms. The relationships between indoor temperature and outdoor temperature of broiler houses and layer houses are different (Fig. S1). In layer houses, temperature is considered to be primarily dependent to the outdoor temperature, while broiler houses' temperature is also related to broilers' body weights. The data for when broilers' body weight is less than 0.5 kg per bird are excluded from the parametrization because a) broilers that are smaller than this size do not contribute significantly to NH3 emissions and b) houses are kept warmer than normal for the smallest chicks compared to birds heavier than 0.5 kg. By excluding these data for small birds, a much better relationship can be found between indoor and outdoor temperatures (Fig. S1), which is also representative of the periods of significant NH3 emissions. In running the AMCLIM-Poultry model for global upscaling, the same relationship from Fig. S1 is applied for all weights of birds, including layers and broilers. These range from chicks to harvested adults and as special conditions are typically applied for chicks. Chicks are typically reared under relatively warm conditions, with the temperature around 32 35°C. However, NH3 emission at this stage is tiny because the nitrogen exerction rate of chicks is low, and litter is typically fresh. For broilers, NH3 emission mostly takes place from the later growing period once exerction rates are larger and litter has built up. Based on the measurements from animal house CA1B (Table S1), the indoor temperature of broiler housing was taken into account (as shown in Fig. S1) for the period in which the body weights exceed a threshold of 0.5 kg.

## 3.1.2 Factors affecting UA hydrolysis rate

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Decomposition of UA from chicken exerctions to TAN is dependent on the temperature, moisture, and pH of the substrate. The maximum estimated breakdown rate is 20 % per day at 35 °C, pH 9.0, and RH 80 % (Elliot and Collins, 1982). The combined influence of three factors is the product of a series of conversion rate functions as expressed by the Eq. (20).

$$5 \quad K_{(T,nH,RH)} = 0.2 \, k_{nH} k_T k_{RH} \tag{20}$$

We used the pH dependence for the range of 5.5-9.0 from the Elliott and Collins (1982) study:

$$k_{pH} = \frac{1.34(9H) - 7.2}{1.34(9) - 7.2} \tag{21}$$

The temperature and RH dependence of UA hydrolysis rate derived from using the AFO monitored data are shown in Fig. 3, where they are compared to functions from Elliott and Collins (1982). The new temperature dependence follows an exponential relationship, and is normalised to the maximum rate at 35 °C:

$$k_T = \frac{\exp^{(0.149(T-273.15)+0.49)}}{\exp^{(0.149(35)+0.49)}} \tag{22}$$

The new RH dependence increases linearly as RH increases, reaching the maximum rate of 1 at RH 80 %:

$$k_{RH} = 0.0124 \, RH = 0.0014$$
 (23)

It should be noted that the RH dependence within the range of RH 0-40 % is extrapolated because there were limited data at these conditions (Fig. 3b).

#### 3.1.3-2 Resistance within chicken houses and site simulations

The inversion derived resistance within chicken houses, R\*, is presented in Figures S2 to S5 (Supplementary Sect. 4); strong daily variations can be seen. The possible relationships of calculated R\* values to temperature and ventilation rate were investigated. This showed no strong correlation with these indoor environmental variables (See Fig. S6 and Fig. S7). We simulated the total NH<sub>3</sub> emissions with various constant R\* values throughout the year and compare the results to the measurements (Fig. S8). A fixed R\* value of  $\sim 16700 \text{ s m}^{-1}$  was found to provide the best result of 1:1 for House A, and  $\sim 14369 \text{ s m}^{-1}$  for House B at NC2B.

Figure 4 and 5 show the simulated indoor NH<sub>3</sub> concentrations and emissions comparing to the measurements by assuming the fixed R\* value of 16700 and 14369 s m<sup>-1</sup>, respectively. Gaps shown in measured concentrations and emissions of NH<sub>3</sub> represent unavailable measurements, while the model was kept running during gaps to produce continuous output. Gaps occurred in measured NH3 concentration and emissions were due to unavailable measurements, while the model was kept running. The model was able to capture the major changes throughout the simulation period. During hot periods of the year, the temperature inside the house was generally higher than cold months, and ventilations rates reached the maximum. High temperature led to large UA hydrolysis to increases the TAN pool, which allows more NH<sub>3</sub> emissions. High ventilation rates accelerated the NH<sub>3</sub> removal from the house, and the indoor concentration of NH<sub>3</sub> decreased. The TAN pool of both houses accumulated and reached approximately 5 kg m<sup>-2</sup>, while the UA pools were relatively low due to the continuous conversion to TAN. Sharp

declines of the UA pools were seen (dates April/09/2008 in House A, June/03/2008 in House B), linked to the chicken houses being empty at these times (as shown by black dash lines) for approximately three weeks. The NH<sub>3</sub> concentrations at the surface were much higher than the NH<sub>3</sub> concentrations of the house atmospheres in both houses. As a result, with sufficient TAN and large difference between surface and air NH<sub>3</sub> concentration, NH<sub>3</sub> emissions in hot-summer months were higher than in winter months. The model overestimated NH<sub>3</sub> emissions from early April to early July and then underestimated the emissions in September for House B. The discrepancies are mainly caused by the use of a fixed housing resistance, R\*. In reality, R\* will vary with the environmental conditions within chicken houses. However, we consider it well justified to use a constant value of R\* in order to keep simple the overall fit of the dataset to the measured emissions, which also simplifies the global application.

#### 3.1.43 Model sensitivity to temperature and relative humidity

To understand the effects of temperature and relative humidity on the NH<sub>3</sub> volatilization in chicken houses, we ran simulations under idealised conditions. We used a configuration (i.e. animal number, house size) the same as the NC2B House A, <u>but but</u> set the temperature and relative humidity to constant values throughout the whole year. A spin-up year run was prior to the experimental simulations.

We tested the NH<sub>3</sub> volatilization rate (Pv) under a domain with temperature range of 15-35 °C and RH range of 20-100 %. Figure 6 shows an overall increasing of Pv from low temperature and RH to high temperature and RH regime. The highest Pv values reaching approximately 56 % were from high temperature and RH simulations. Figure 7a shows that the Pv rates increase as temperature increases, and Fig. 7b also shows that the Pv rates increase as RH increases, but drop after RH exceeds 90 %.

## 20 3.2 Site simulations for land spreading

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We ran a set of simple site experiments for land spreading to quantify the NH<sub>3</sub> volatilization under different environmental conditions. The model configurations of these simulations are given in detail in the Supplementary Sect. 5. We compare the model results with reported measurements from five experimental studies (Lau et al., 2008; Marshall et al., 1998; Miola et al., 2014; Rodhe and Karlsson, 2002; Sharpe et al., 2004). There are three groups of comparisons that represent different simulation and measurement duration: 7, 14 and 21 days, respectively.

As shown in Fig. 8, the simulated percentage of nitrogen excreted that is volatilized as NH<sub>3</sub> (Pv, %) increases as temperature increases, because of the faster UA hydrolysis rate in hotter conditions. The shaded areas illustrate ranges of Pv from simulations that use different RH values ranging from 20 to 100 %, while the solid lines represent the mean Pv rate for the range of RH values for each simulation period (7, 14, 21 days). Compared with the experimental studies, the model application underestimates NH<sub>3</sub> volatilization for the 21 days simulation and overestimates for the 14 days simulation. However, it is evident that these experimental studies also show large variations, which we expect is especially due to meteorological variation within and between the experimental studies, such as rainfall or windy conditions. For example, at a mean temperature of around 26 °C Sharpe et al. (2004) reported Pv of 23 % and 5 %, respectively. The latter value was caused by a rain event taking place two days after application, explaining why the latter point appears low on Fig. 8 where the simulations are based on rain free conditions. Overall, the model provides Pv rates that fall within the range between 0.5x to 2x compared to the measurements. It should be noted that this is a very simple model experiment because the published experimental studies do

not always fully describe environmental conditions, which limits the extent to which features of the AMCLIM-Poultry can be applied for comparison with the measured datasets.

## 3.23 NH<sub>3</sub> emission from global chicken housing

We used the polynomial fits shown in Fig. S1 and the constant R\* values of 16700 s m<sup>-1</sup> as representative of all chicken houses for the simulation of global emissions. The estimate of NH<sub>3</sub> emission from global chicken housing in 2010 was 2185.5 Gg2.0 Tg N. This includes 1374.71.3 Gg Tg N emissions from broilers and 810.80.7 TGg N from layers. Figure 8-9 shows high emissions in Europe, India, China and Southeast Asia, with emission hotspots in eastern US, and the eastern part of South America. The total amount of nitrogen from chicken excretion was 9017.1 Gg9.0 Tg N in 2010. The percentage of nitrogen excreted that is volatilized as NH<sub>3</sub> (Pv, %)volatilization rate, Pv, was estimated at 24.222 % overall for all NH<sub>3</sub> emissions from chicken housing globally. The value of Pv for chicken housing was high across the tropics, reaching approximately 35 % (Fig. 8b9b). Regions with high NH<sub>3</sub> emission mostly show high NH<sub>3</sub> volatilization rates, especially in regions such as east China, Southeast Asia, and east US. As the Pv value normalizes for chicken numbers, it more clearly shows the influence of climate than total NH<sub>3</sub> emissions. Figure 8b-9b shows very small Pv values in dry areas (Sahara, Australia, Arabian peninsula Arabian Peninsula, Patagonia, Central Asia, western North America, illustrating low humidity in these areas is estimated to limit UA hydrolysis, with the converse in humid areas (Amazonia, central Africa, south east Asia, etc).

# 3.34 NH3 emission from global chicken manure spreading

## 3.34.1 NH<sub>3</sub> emission from chicken manure application for crops

For the year 2010, the NH<sub>3</sub> emission from chicken manure application for crops was 2582.32.7 TGg N, with the Pv value representing 37.839 % of the total nitrogen application to land of 6827.0 Gg7.0 Tg N. The nitrogen considered to be left untreated according to Sect. 2.4.3 was 4.6less than 50 Gg, which is only a small fraction compare to the amount of nitrogen applied to land. From simulations in this study, over 75 % of the NH<sub>3</sub> emissions were from applications for the major 6 crops specified in Sect. 2.4.3, while the rest were from applications for other crops (Table S2 in Supplementary Sect. 7). Among the 6 crops, maize fertilising contributed to the highest emission of 643676.4 Gg N, which is more than approximately 1/3 of the total amount. Fertilising rice and wheat also led to 601.4641.2 and 520.3542.7 Gg N of emissions, respectively. Compared with maize, rice and wheat, crops of barley, potato and sugar beet had much smaller emissions due to lower estimated total application of chicken manure to these crops (reflecting their smaller cropping areas and the chicken distribution). The NH<sub>3</sub> volatilization of crops all six crop types exceeded 34.35 % (Table S2). The application for rice resulted in the highest Pv of over 42.043 %, (reflecting the warm and moist climate of rice cropping), while the application for barley and sugar beet had the lowest Pv values of 34.536 % (reflecting its distribution in cooler temperate climates).

The geographical distribution of NH<sub>3</sub> emissions from chicken manure application is presented in Fig. 9a10a. Similar to the chicken housing, high emission can be seen in Europe, eastern Middle East and south India, while extremely large NH<sub>3</sub> emission exceeded 10 Gg N yr<sup>-1</sup> over eastern and central part of China and south east Asia, with hotspots in south eastern US, Mexico and eastern South America. These hotspots reflect a combination of high chicken populations and high P<sub>V</sub> values. Areas of the lowest P<sub>V</sub> are associated with cropping areas having the lowest rainfall, including west central North America,

southern Africa and central Asia. Areas estimated to have no significant arable cropping (i.e., desert, boreal and tundra) are shown white in Fig. 910.

### 3.34.2 NH<sub>3</sub> emission from backyard chicken

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The global NH<sub>3</sub> emission from backyard chicken in 2010 was estimated at 714.5 Gg0.7 Tg N from a total excreted nitrogen of 2178.32.2 TGg. Backyard chicken density showed a different distribution compared with broilers and layers (Fig. S10 in Supplementary Sect. 8). This reflects the assessment in the FAO database that backyard chickens are not kept in developed countries including Canada, United States of America, west Europe, Australia and New Zealand, where all chicken are allocated to housed systems. The FAO database estimates that most backyard chicken occur in developing regions, such as the northern India and Africa. Geographically, the highest emission from backyard chicken are here estimated to occur in Ukraine, south and south-east Asia, with high emissions in east coastal regions of South America and the southern part of West Africa. Figure 10b-11b illustrates the geographic distribution of the percentage nitrogen volatilized (Pv). The volatilization rates of vast majority of Asia were less than 24 %, while the tropics including South Asia had higher Pv rates that reach 36 %. Possible reasons for the different distribution of Pv for backyard birds as compared with manure application to crops are discussed in Sect. 4.2.

## 5 3.45 Annual NH<sub>3</sub> emission inventory ffor rom global chicken farming

The estimated NH<sub>3</sub> emissions based on 2010 are summarised in Table 1, and the geographic distribution is presented in Fig. 4+12. Overall, the total emission from global chicken farming was 5482.35.5 TGg N yr<sup>-1</sup>. Practice related to broilers and layers including housing and manure application to crops contributed 2185.52.2 and 2582.32.7 TGg N NH<sub>3</sub> emissions, respectively, and backyard chicken manure caused 714.5 Gg0.7 Tg N emissions. Regions with high NH<sub>3</sub> emissions were across Europe, India, and part of China, with hot spots occurred in East US and Eastern South America. The distribution of Pv values reflects the combined effect of how environmental differences lead to variations in emissions from chicken housing, manure spreading to arable land and from backyard birds.

Figure 12\_13 shows the NH<sub>3</sub> emissions from the three main components for chicken (housing, crops, backyard) and <u>summarizes</u> the latitudinal difference in percentage volatilized the corresponding volatilization for 5 latitudinal bands. The highest emission was between 20 —and 40 °N, reaching a total NH<sub>3</sub> emission of 2540.8 Gg2.5 Tg N. The lowest emission was 317.2 Gg0.3 Tg N between 20 —and 40 °S. Manure application to crops was the largest fraction of NH<sub>3</sub> emissions in the northern hemisphere, and its volatilization to NH<sub>3</sub> was the highest among the three categories across the globe, exceeding 35 %. The NH<sub>3</sub> volatilizations of housing and backyard chicken were comparable, ranging between 20 % to 30 % of the total emission. Figure 12 summarizes the latitudinal difference in percentage volatilized. The smaller degree of variation reflects the complex way in which water availability, humidity and temperature interaction to affect the overall percentage of nitrogen volatilized, as illustrated by the maps.

Figure 13a-14a shows the monthly NH<sub>3</sub> emissions from each sector. Highest emissions of over 600 Gg0.6 Tg N were estimated for April and August, while lowest estimated emissions were in November, December and January. This shows how the seasonal differences are larger for NH<sub>3</sub> emissions from manure application to crops than from animal houses, which is a result of both the climatic effects, and the temporal distribution of manure application according to the start of the main cropping seasons. From Fig. 13b14b, the NH<sub>3</sub> volatilization from backyard chicken excretion varied more throughout the year than for

housing (linked to larger variations in temperature and water availability). Emissions from backyard birds were higher than housing from April to August, with the largest difference in July, and were lower than housing from September to March. The highest estimated rate was 65\_4% in July and lowest rate was 12\_2% in January. The volatilization rates of housing showed smaller variations, with P<sub>V</sub> values mostly over 20 %, with the highest rate of 30.928 % occurring in August. It is worth noting that volatilization rates of manure land spreading are not presented in the figure because simple monthly values do not reflect the true volatilization rate. Nitrogen being applied in the agricultural month will cause NH<sub>3</sub> emission in the following months when no application practices take place.

#### 4 Discussion

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## 4.1 Parametrisations Model parameterization for chicken housing

#### 4.1.1 Indoor environmental conditions of chicken houses

Meteorological conditions affect the NH<sub>3</sub>-emissions from chicken housing indirectly by influencing the indoor environmental conditions, which is crucial in affecting NH<sub>3</sub>-volatilization. At high temperatures, the ventilation rate is increased to cool down the house, keeping the inside temperature close to the reference value. When ventilation systems reach their maximum flows, indoor temperature would continue to rise above the reference. Increasing ventilation rates help minimize temperature increases, increase water evaporation of the house, and reduce the moisture associated with chicken excretion. Theoretically in warm dry conditions, net NH<sub>3</sub>-emission tends to decline because of the less efficient UA hydrolysis. By contrast, in humid conditions, increased ventilation of chicken houses under warmer conditions is estimated to increase NH<sub>3</sub>-emissions in the model, as UA hydrolysis is favoured and NH<sub>3</sub> are quickly removed from the houses to the atmosphere.

It is worth noting that management for broiler rearing is different from layers. The growth period of which broilers from chicks to adults is approximately 6-8 weeks. At initial stage, the house is heated to keep the inside temperature up to 32-35 °C, allowing the chicks to grow under a warm and comfortable condition. As the birds are growing stronger and gaining weight, the indoor temperature is decreased. Once the adult birds gain enough weight, they are removed from the house. The house is then empty for the next 3-7 days until another flock is settled in, and the heating system is turned off. In comparison, egg layers are kept longer in houses, which normally lasts for over 2 years. The indoor temperature of a layer house is controlled, as far as possible, within a referenced range throughout the year. The manure management also varies. According to the AFO's dataset, broiler houses in the US are cleaned after every 3-4 flocks, and the exerction with litter or bedding materials removed, while layer houses are usually designed to have multiple floors, allowing the litter to be collected and removed at the lower floor by conveyor belts. These differences have implications for NH<sub>2</sub>-emissions between broiler and layer systems, the most important one being the need to recognize the cycles of temperature and humidity as these affect NH<sub>2</sub>-emissions from broilers. Even if litter is not cleared out after removing grown broilers, it is anticipated that new bedding material will be added, therefore covering the old litter, so that emissions are mainly related to the excretion of each flock. Since most emissions are associated with older broilers, this has allowed the simplification (Sect. 3.1.1), that the relationship between indoor and outdoor temperatures is based on periods where birds are >0.5 kg. While the relationship between indoor and outdoor temperatures applied here is based on the US EPA experimental farms, access to such datasets for other climates would be useful to extend and improve the parametrization.

## 4.1.2 Comparison between the empirical equations for UA hydrolysis in chicken housing

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Figure 3 shows the parameterizations for UA hydrolysis in chicken houses from this studythat is derived from AFO's measurements and is taken from the Elliott and Collins (1982) study. The temperature dependences are comparable in that both studies suggest an exponential correlation between the Factor T and indoor temperature. Overall, the Factor T derived from using the AFOs monitored data in this study was slightly larger than that from Elliott and Collins (1982). Within the temperature range of 18 to 28 °C, the UA hydrolysis rate approximately doubled every 5 °C, and an increasing 10 °C led to more rapid hydrolysis rate by a factor of 4.4 and 5.2 based on the two studies, respectively. In contrast, the RH dependences were more different between the two studies. The new parameterization suggests a linearly decline of Factor RH as RH decreases below 80 %, so that the magnitudes of Factor RH are much larger compared with Elliot and Collins (1982). When RH is below 40 %, the Factor RH for the present study was obtained from extrapolation due to the lack of measurement from the AFOs dataset.

The results of global housing simulations by using two parameterizations are presented in Fig. \( \frac{8-9}{2} \) (using RH parametrization from Elliot and Collins, 1982) and Fig. S9 (using the new RH parametrization based on Fig. 3 from the monitored AFOs). The annual NH<sub>3</sub> emissions from housing in 2010 were estimated at 3312.4 Gg3.0 Tg N based on the new parameterization (from the monitored AFOs), giving 51.60 % higher emissions than the estimates of 2185.5-.0 TGg N using the equations of from Elliott and Collins (1982). In principle, warmer and wetter conditions lead to an increase in Pv. Increasing temperature accelerates the formation of TAN and increases the surface concentration of NH<sub>3</sub>, and the hydrolysis of UA is enhanced under high moisture environments. The temperature inside chicken houses in the AMCLIM-Poultry model is assumed to be controlled, especially the houses in cold climate regions, where sufficient heating is assumed to be used to maintain healthy environments. Therefore, the variations of housing temperature were not as significant as the outdoor temperatures. On the other hand Meanwhile, the houses prevent the rain getting in, so the hydrolysis of UA and aqueous NH<sub>3</sub> concentration are solely restricted by the water content of the excretion, which is a function of RH. As a result, RH becomes the foremost factor that determined the NH<sub>3</sub> emissions by affecting the water availability of the system. It is notable that large differences between the two sets of global simulations (as shown in Fig. 8-9 and Fig. S9 in Supplementary Sect. 6) occurred in dry regions, such as Northern Africa, the Middle East, and Western Australia. Compared with the results of using the Elliott and Collins equations, the new parameterization suggests much higher NH<sub>3</sub> volatilization in dry places. The substantial difference between the model simulations using the two RH parametrizations indicate the need for further data on this relationship. Additional measurement datasets including both temperature and RH measurements, and representing a wider range of environmental conditions, would help to strengthen and extend the relationships observed. The RH dependency of UA hydrolysis from Elliot and Collins (1982) was used for outdoor simulations that includes land spreading and backyard chicken, which has been previously tested and found to provide robust estimates from the GUANO model (Riddick et al., 2017).

It must also be recognized that both the RH parametrizations shown in Fig. 3b have limitations. A more accurate parameterization of RH dependence might fall in the area between two curves in Fig. 3b. It can be seen from Fig. 4c and Fig. 5c that the TAN pool of each chicken house increased continuously throughout the simulation period rather than remaining approximately constant at some points. This indicates that the TAN produced exceeded the loss through NH<sub>3</sub> emission, which is against the assumption that the production of TAN is equivalent to the NH<sub>3</sub> emission. It is possible that this the new RH dependence overestimated the rate of UA hydrolysis. Meanwhile, from the Fig. S4 and Fig. S5, by using Elliott and Collins's parameterization for RH dependence of UA hydrolysis equation, the modelled indoor concentration of NH<sub>3</sub> was much lower than the measurements during the starting period of simulations. This was caused by the indicates an insufficient TAN pool

that limited the emissions. Therefore, Elliott and Collin's parameterization probably underestimated the TAN production from UA hydrolysis, especially when each nitrogen pool was limited. In addition to the need for further datasets that relate NH<sub>3</sub> emissions from housed chicken to both indoor temperature and relative humidity, parallel measurements of the water, UA and TAN content and pH of different litter layers would be helpful to improve future parametrization.

#### 5 4.1.3 The NH<sub>3</sub>-transfer resistance of chicken houses

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The inversion derived resistance within the chicken houses, R\* at NC2B typically ranged from 10000 s m<sup>-1</sup>up to 50000 s m<sup>-1</sup> with strong variations. According to Pinder et al. (2004), from a dairy manure storage sub model with parameter tuning, the surface resistances of crust with wheat straw ranged between 0.1 to 0.4 day m<sup>-1</sup>, which corresponds to 8640 s m<sup>-1</sup> to 34560 s m<sup>-1</sup>. As no obvious correlation between R\* and environmental factors was found, it remains unclear that by which parameters the R\* are affected. Based on the conditions of two chicken houses at NC2B, the sensitivity test of using constant R\* value to simulate 1 year NH2 emissions suggested that the R\* values led to the best agreement with the measurements were 16700 and 14369 s m<sup>-1</sup>, respectively. It is worth noting that the best fit R\* values for each house are smaller than the mean or median values of the inversion derived R\* values. This indicates that a relatively small R\* value leads to a good approximation of the fraction of TAN pool being depleted through NH2-emission, while R\* becomes less effective on restricting NH2-emissions as its value increases. For the House A, change of R\* from 8350 to 33400 s m+caused the ratio (of simulated to measured NH<sub>3</sub> emission) decrease from 1.24 to 0.75. Likewise, changing R\* within the House B from 7185 to 28740 led to the ratio ranged between 1.25 to 0.73. The varying of R\* value by a factor of 2× resulted the total NH<sub>3</sub>-emission for a whole year period changing approximately 25 %. This implies that under current housing conditions, the total annual NH<sub>2</sub> emission is not strongly influenced by the resistance within the houses. Instead, resistance plays more crucial role in affecting the short term emissions. Large resistance limits the emission initially, but leads to the TAN accumulation to allow larger emissions at a later point in time, therefore reducing the overall sensitivity to R\* for annual timescales.

# 4.1.24 Implications for the idealised simulations

As shown in Fig. 6 and Fig. 7, it can be seen from dry simulations (i.e., without precipitation) <u>under idealised conditions for a whole year run</u>-that the annual mean P<sub>V</sub> was relatively small and can drop to approximately zero when temperature is low. It indicates that the UA hydrolysis is hardly to take place. In contrast, the P<sub>V</sub> were much higher in hot and wet regimes, reflecting an effective hydrolysis of UA. It is notable that the P<sub>V</sub> declines at very high RH levels using the new RH parametrization. This is mainly because the UA hydrolysis is considered to be optimum at 80 % and higher RH, but the TAN concentration becomes lower as the excretion contains more water when the ambient environment is humid, thereby providing a "diluting" effect.

From Fig. 7a, the  $P_V$  rate is seen to grow exponentially as a function of temperature for the 20 % RH simulations. It is similar to the impact of temperature on UA hydrolysis and also the Henry's Law relationship. Conversely, for a humid environment with RH at 100 %, there is a smaller increase of  $P_V$ , showing a logarithmic-like trend. These differences are consistent with different amounts of TAN under the two cases. When there is sufficient TAN produced from the UA hydrolysis, the resistance can become the key limiting factor to emission from the system. Conversely, in low-humidity environments, as the UA hydrolysis is limited, the produced TAN is readily removed through the atmospheric release of NH<sub>3</sub>, with total emission limited by the UA hydrolysis rate. Therefore, the rise of temperature under dry conditions provides a larger increase in NH<sub>3</sub> emissions.

From Fig. 7b, it is worth noting that the decrease of P<sub>V</sub> occurs when the RH slightly exceeds 90 % rather than 80 %. A more obvious sharp decline can be seen from the 15 °C simulations. As discussed, there is a "diluting" effect on the TAN

concentration when the RH is over a certain level. The possible reason why this turning point does not occur at the 80 % RH where is the factor RH reaches the optimum can be summarised as follows. The  $P_V$  rates in these simulations represent the integral of a whole year. The "diluting" more water to dissolve TAN at high RH affects the instantaneous emission without changing the amount of TAN pool. Low emissions in the earlier stage can therefore cause a larger emission potential in the later stage due to accumulation of TAN.

The overall implication of these idealized simulations is to demonstrate the close interplay between water availability and temperature, where temperature always increases volatilization (partitioning in favour of the gas phase), whereas a small amount of water is needed to facilitate UA hydrolysis, increasing NH<sub>3</sub> emissions, while excess water availability dilutes the TAN pool, thereby reducing NH<sub>3</sub> emissions. These same principles also apply for emissions from manure application to crops and for backyard birds, where precipitation and run-off become more important.

#### 4.2 Spatial and temporal variations of NH<sub>3</sub> emission

The NH<sub>3</sub> emission from chicken agriculture differs substantially across regions, both because of different chicken number distributions (Supplementary Fig. S10), as this affects total nitrogen excretion, and because of different volatilization rates, as shown by the Pv values. The largest NH<sub>3</sub> emission is calculated for regions between 20 ~ 40 °N, which corresponds to the highest chicken density and associated manure application to land. The animal number and the amount of nitrogen from excretion have a first order effect on the magnitude of emissions. Considering the Pv, the most significant spatial variations relate to emissions from manure spreading and backyard chicken, with less spatial variation in Pv for housed birds as the indoor conditions are considered to be largely controlled. Considering the variations in Pv, there is most estimated variation in NH<sub>3</sub> volatilization of manure spreading and backyard. The Pv rates of backyard chicken excretion were much lower in China and Southeast Asia by comparison with manure land application, because the wash off is a major loss of nitrogen pools in these regions, especially during non-cropping periods when chicken manure is not applied to land (according to our model approach), while backyard birds lead to outdoor NH<sub>3</sub> emissions all year round (including during non-cropping periods with high precipitation).

It should be noted that from the northern India to Tibet, the P<sub>V</sub> rate declines sharply from 40 % to below 6 % from all categories. This indicates that a sudden change from hot and wet conditions to cold and dry conditions causes the volatilization rate drops dramatically in Tibet compared with India. This example clearly illustrates how the fraction of nitrogen volatilised as NH<sub>3</sub> is strongly linked to meteorological and related environmental conditions.

The AMCLIM-Poultry simulations also showed strong seasonal variations of NH<sub>3</sub> emissions from manure land spreading and backyard chicken excretion. The seasonal distributions (as illustrated by Fig. 1314) were caused by changes in meteorological conditions, with high NH<sub>3</sub> emissions in summer due to the high temperature influencing NH<sub>3</sub> emissions from housing and backyard birds. Even larger seasonal differences are seen in the modelled emission estimates for land application of manure, because this combines both the direct effects of environmental variation (temperature and water effect on P<sub>V</sub>) with seasonal differences in the estimated timing of manure application to land. Paulot et al. (2014) found that maximum NH<sub>3</sub> emissions from manure fertilising can occur from April to September depending on the local management. For example, they found that emission peaks in spring occurred in Europe, while summer emission peaks occurred in part of the US and China. These differences reflect a combination of agricultural timing and the meteorological/environmental drivers (Hertel et al., 2011). Riddick et al. (2016) also showed the maximum emissions usually occur in April-June or July-September. The findings in

present study are broadly consistent and demonstrate for the first time on a global scale how emissions from managed poultry (chicken) are dependent on both short-term meteorology and long-term regional climatic differences. Contrary to manure spreading and backyard birds, the seasonal variations of NH<sub>3</sub> emissions from chicken housing were much smaller due to the partly controlled environment and the assumed absence of precipitation/run-off within the houses.

## 5 4.3 Comparison with other inventories and models

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We compared the results from the AMCLIM-Poultry model to three other (model-based) studies/reports from Denmark, Netherlands and United Kingdom, respectively. The Danish IDA model (Albrektsen et al., 2017) and the UK NARSES model (Misselbrook et al., 2011) provided 2010 emission data, and the NEMA model (Velthof et al., 2012) from Netherlands estimate emissions in 2009 (see Table 2). All these studies report emissions from poultry rather than chicken. It has been clearly stated that the input used in the AMCLIM-Poultry from the GLEAM model used here are chicken data, which excluded other poultry such as turkeys, ducks etc. Therefore, we can see that the excreted nitrogen from the GLEAM model (GLEAM FAO, 2018) is generally smaller than other individual studies. For housing, the AMCLIM model shows similar estimates of NH<sub>3</sub> emissions to the other models. The housing emissions from this study are smaller than the local models in Denmark and Netherlands, partly due to the smaller total excreted nitrogen from animals. However, the AMCLIM model suggests larger emissions from land spreading for Netherlands and the UK (spreading-derived emissions are not available from the IDA model), especially in Netherlands where the difference between the two estimates reaches 8x. This is probably due to the different schemes or assumptions for land spreading practices, e.g., deep injection of manure, in different models. The Pv rates, which indicate the fraction of nitrogen that is emitted as NH<sub>3</sub> are comparable from all models for the housing sector. The AMCLIM model suggests that the Pv rates do not vary significantly between these countries because the indoor conditions are largely controlled and in similar climates, which leads to small variations in house environments.

In addition, we also compared our results with existing emission factors (EFs). On a global average, the AMLCIM model estimated that the EFs for broiler and layer housing are 0.13 and 0.10 kg N animal<sup>-1</sup> yr<sup>-1</sup>, respectively. Combining with emissions from land application, the total EFs are 0.30 and 0.27 kg N animal<sup>-1</sup> yr<sup>-1</sup> for broilers and layers, and the EF for backyard chicken is 0.19 kg N animal<sup>-1</sup> yr<sup>-1</sup>. Regionally, the AMCLIM model estimates that the UK have EFs of 0.13 (0.11–0.14) kg N animal<sup>-1</sup> yr<sup>-1</sup> for chicken housing and 0.30 (0.12–0.33) kg N animal<sup>-1</sup> yr<sup>-1</sup> for the total emission, compared to 0.10 (0.06-0.15) for housing and 0.22 (0.15–0.30) for the total EF reviewed by Sutton et al (1995a). For Europe, the EFs estimated by the AMCLIM model are 0.10 (0.01-0.16) and 0.09 (0.01-0.15) kg N animal<sup>-1</sup> yr<sup>-1</sup> for broiler and layer housing, and 0.15 (0.01-0.28) kg N animal<sup>-1</sup> yr<sup>-1</sup> for the followed land application. In comparison, according to the EMEP/EEA (2019), EFs are 0.16 to 0.32 and 0.15 kg N animal<sup>-1</sup> yr<sup>-1</sup> for layer housing and consequent manure application, while EFs for broiler housing and manure application are 0.13 and 0.04 kg N animal<sup>-1</sup> yr<sup>-1</sup>.

## 4.3-4 Uncertainty and limitations

There is substantial uncertainty in modelling NH<sub>3</sub> emission from livestock farming. Here, we focus on discussing the uncertainty related to model parameterizations. The model parameters may influence the emissions interactively with non-linear consequences. We find that it is helpful to conduct sensitivity analysis by simulating the effect of changes in parameters on NH<sub>3</sub> emissions. By doing this, we are able to indicate the ranges of uncertainty and also to highlight which parameters are most important and need to be further investigated. Based on prior test, we find that indoor resistance R\*, manure pH, runoff coefficient and amount of N excreted are most important and examine these in the sensitivity tests, with results summarised in

Table 3. In addition, the uncertainty arising from the parameterization of UA hydrolysis is represented by the differences between Fig. 9 and Fig. S9.

It is worth noting that the ranges of the parameters are based on expert judgement. Indoor resistance and runoff coefficient are considered to be uncertain by a factor of 2, with manure pH uncertain by  $\pm 1$ , which corresponds to a factor of 10 for hydrogen ions concentrations. The nitrogen excretion rate is considered to have an uncertainty of 10 %. The global simulation of housing driven by varying indoor resistance values shows that 2x higher R\* leads to NH<sub>3</sub> emission decrease by approximately 31 %, and 2x lower R\* leads to 27 % higher emissions, which is similar to the result at the site scale (see Fig. S8). The R\* values directly influence the magnitude of housing emissions, but only to a limited extent. The R\* values also impact NH<sub>3</sub> emissions from land spreading of chicken manure by limit the available amount of nitrogen that is applied to land. In total, doubling R\* <u>leads</u> to a reduction of NH<sub>3</sub> emissions by 6.4 %, and half R\* leads to an increase of emissions by 8.5 %. The manure pH, which affects the hydrolysis rate of UA and the chemical equilibria between NH<sub>4</sub><sup>+</sup> and gaseous NH<sub>3</sub>, is found to have positive effect on NH<sub>3</sub> emissions that emissions tend to increase as pH increases. We find that increasing pH from 8.5 to 9.5 causes annual NH<sub>3</sub> emission to increase by 5.8 %, while a decrease of pH to 7.5 leads to a decline of emission by 15.9 %. The runoff coefficient was set to be 1 % mm<sup>-1</sup> for nitrogen pools in the model (Riddick et al., 2017). By doubling the runoff coefficient, 15 the NH<sub>3</sub> emissions decrease by 11.8 %, while decreasing the coefficient to half lead to emissions increase by 16.5 %. It should be noted that among these parameters, changing the manure pH has influences on both housing emissions (from broiler and layer housing) and outdoor emissions (spreading of broiler and layer manure; backyard chicken manure). The runoff coefficient only affects the outdoor emissions, while indoor resistances limit housing emissions directly, but also have impacts on consequent outdoor emissions. Smaller NH3 emissions from housing indicate a larger potential for outdoor release during the spreading stages under the same farming practices. Conversely, higher housing emissions lead to smaller consequent emission 20 from land application. Concerning the nitrogen excretion rate from chicken, find that a 10 % of variation leads to an annual NH<sub>3</sub> emission change of approximately 12 %. The change in NH<sub>3</sub> emission is not proportional to the nitrogen input because of non-linear interactions in the model, e.g., an increase in nitrogen input by 10 % may only lead NH3 emissions to increase by a negligible amount in regions with heavy rainfall. Combining these ranges and taking the base run result as the "best estimate", the overall expected uncertainty of NH<sub>3</sub> emissions from global chicken farming is 1.2 Tg N yr<sup>-1</sup>, with component 25 uncertainties of housing, land spreading and backyard chicken are 0.6, 0.5 and 0.2 Tg N yr<sup>-1</sup>, respectively. Detailed estimates are described in Supplementary Sect. 9.

Future directions of the study include a) a better parameterization for UA hydrolysis, b) developing an interactive scheme for soil interactions, which allows to simulate soil pH dynamically and relevant soil processes such as absorption of TAN, c) incorporate more detailed pathways for nitrogen flows, such as nitrification and leaching, and canopy recapture, and d) a better representation of human management based on statistical data or national and international survey.

# 4.3.1 Simulation of emissions from chicken housing and storage

For simulating NH<sub>3</sub> emissions from chicken housing, the largest uncertainties are mainly associated with the model parameterizations linked to temperature (T) and relative humidity (RH). As all the measurements used were from the US chicken farms, the modelled values of the RH and T parametrizations (Fig. 3) provide only a first estimate to represent variation in climatic conditions on a global scale.

According to our methodology, the parametrization of Fig. 3 is applied to all housed chicken across different climates. However, it is possible that a substantial number of chicken houses are not climate controlled in any way. For example, in tropical countries intensively managed chicken houses may not have any (or only limited) heating and ventilation systems. In this context, a larger fraction of chicken houses may be naturally ventilated throughout the year because cold days are usually very rare. In this case, the temperature inside the chicken house would be simply 2.5°C above the outdoor temperature due to the heat generated by the chicken themselves, with airflow rates are related to natural wind speed. In such a naturally ventilated situation, there may be no steady state between the NH<sub>3</sub> emission from the surface and the removal through ventilation. With the availability of appropriate data, such altered ventilation regimes could easily be included in the AMCLIM Poultry model, and would be expected to show an even larger temperature dependence for chicken housing emissions than estimated using the present parametrization.

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Second, due to lack of other data, the new parametrisation for UA hydrolysis is primarily derived from specific chicken houses, under US conditions. These chicken houses had explicit clean out dates for the dataset, which allows the model to be run under a specific initial condition that each nitrogen pool is empty at the beginning. It remains unclear how the model will perform with the new parameterizations for chicken houses that are already loaded with manure. Meanwhile, the equations given by the previous study of Elliot and Collins (1982) resulted in a large discrepancy between the modelled values and measured data during the earlier stage of the simulations. It is evident that there is a need for further experimental datasets for a wider range of climate conditions, including all available indicators (NH<sub>3</sub> emissions data and ventilation data accompanied by both temperature and relative humidity, stocking timing and ideally data on manure characteristics). From a modelling perspective, a possible approach of introducing different vertical layers into chicken litter could be useful to investigate the effect of adding fresh bedding onto old, deep litter. However, the additional complexity would need to be judged against the potential benefits.

Third, as the litter in the houses are not subject to precipitation or evaporation, the water amount of the system is calculated from the excretion mass and the equilibrium moisture depending on the RH and temperature. The model is not able to simulate the evaporation from the litter in the chicken house. Therefore, the litter moisture is assumed to be at equilibrium. The weakness of this method is that the initial water within the excretion is not accounted for, which might cause uncertainty.

Fourth, the indoor resistance for NH<sub>3</sub> transfer within the chicken houses (R\*) needs further investigation. Pinder et al. (2004) applied indoor resistance with dairy houses that were tuned as a function of temperature. English et al. (1980) developed a series of mass transport coefficients given as a function of wind velocity. As there were no specific correlations between environmental factors and the resistance found in this study, we used a constant value in the simulations rather than parameterised. While this provides a significant uncertainty for short term (e.g., daily) fluctuations in NH<sub>2</sub> emissions, model feedback reduces the sensitivity over annual timescales, as slow emission earlier (associated with high R\*) allows increased emission at a later stage, and *vice versa*. While measurement approaches to estimate R\* would be welcome (e.g., using water vapour loss from wetted surfaces or other tracers), the value of R\* is therefore not considered the largest uncertainty in the seasonal and annual simulations.

The version of AMCLIM-Poultry applied here does not explicitly treat NH<sub>3</sub>-emission from stored chicken manure as a separate step. Emissions from in-house storage of manure are considered as part of the housing calculations, while losses in the field are linked to conditions for land application of manure. For the purpose of the model, which focuses primarily on assessing the climatic dependence on NH<sub>3</sub>-emissions, it is assumed that the climatic dependence of emissions from any storage of chicken

manure outside of animal houses and prior to manure spreading follows the same climatic or intermediate climatic dependence between housing and manure spreading. Future work may consider the case to include an additional AMCLIM module for outdoor storage of chicken manure, where the main uncertainties concern: a) providing a basis to estimate the appropriate outdoor manure storage time according to climate and regional practice, b) providing a basis to consider depth and surface area of stored manure, c) providing a basis to estimate the fraction of manure that is stored outside or under cover. Although the input assumptions are expected to introduce substantial uncertainty, the actual simulations would represent a straightforward extension of the AMCLIM-Poultry approach.

#### 4.3.2 Simulation of emissions from agricultural land

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Outdoor NH<sub>2</sub> emission from chicken manure consists of two parts: manure fertiliser from broilers and layers applied for crops 10 and backyard chicken excretions left on land and pastures. A major uncertainty in the simulations is the amount of nitrogen input from chicken manure to crops. There are multiple management options for chicken manure, including composting, burning, and various storage (FAO, 2018). The amount of nitrogen applied for individual crops as input to the model might be overestimated due to the simple comparison method in this study. Meanwhile, as simulations for both processes were run under natural environments, the following parameterizations incorporated in the model also cause uncertainty.

15 First, the pH of the substrate can greatly affect the NH<sub>3</sub> volatilization by influencing the UA hydrolysis and the TAN partitions that determines the surface concentration of gas phase NH<sub>3</sub>. The pH of the system is dependent to chicken manure pH and soil pH. The chicken manure pH is mostly alkaline, with reported measurements in a range of 7.23 to 9.1 (Sommer and Hutchings, 2001). For the soil pH, there are spatial variations in the geographical distribution. The typical values depending on the crop types range between 5.8 to 7.0, which is usually lower than the pH of chicken manure (Riddick et al., 2016). A major difficulty 20 in determining the pH of the system is because the hydrolysis of UA and NH<sub>2</sub>-production can change the soil pH. The NH<sub>4</sub><sup>+</sup> produced by the decomposition of UA disassociates to form gaseous NH<sub>2</sub>, resulting in H<sup>+</sup>consumption, resulting a sharp increase of soil pH in the initial period and then decrease again in the following days (Chantigny et al., 2004). Móring et al. (2016) proposed a dynamic scheme for simulating soil pH in a field scale model and had a reasonable approximation against measurement. In a following study (Móring et al., 2017), it suggested that a fixed value for soil pH can be used in the modelling 25 of NH<sub>3</sub> emissions in large scales, but the value is uncertain and can differ across regions. Due to the complexity of determining precise pH, a constant value of 8.5 characteristic for solid chicken manure (Elliot and Collins, 1982; Riddick et al., 2017) is used for simulations. While the assumption of the high pH value results in more rapid UA hydrolysis and higher surface concentration of gas phase NH3, leading to more emissions, the present approach was found to agree well with the measured NH<sub>3</sub> emissions for housed chicken and is consistent with the approach validated by Riddick et al. (2018) for wild seabird emissions across different climates...

Second, nitrogen pools including UA and TAN are determined by source and loss, while one of the major loss of nitrogen in land spreading simulations is through run off. The model used a relatively simple approach to calculate the run off. A coefficient multiplied by the amount of each N containing component. The coefficient is a product of two variables, a washoff factor and the water available for wash-off. The wash-off factor was set to 1 % mm<sup>-1</sup> rain for run-off of UA and TAN and 0.5 mm<sup>+</sup>rain for run-off of manure based on the study of Blackall (2004). The available water equals to the total amount of water excluding the water absorbed by the manure that is simply assumed to be twice as the excretion. Although similar parameterization has been validated by the site measurements for seabird colonies (Riddick et al., 2017), there is potential to develop more sophisticated approaches that might be better adapted to simulate emissions from chicken globally.

Third, the model estimated the NH<sub>3</sub> emission without considering the deposition of NH<sub>3</sub> onto the vegetation. Based on previous studies, a large fraction of NH<sub>3</sub> emitted from the surface TAN pool is considered to be captured by vegetation, which could reach 75 % in the case of outdoor bird excreta under a vegetation canopy (Riddick et al., 2016). From the Bouwman et al. (1997) study, plant recapture of NH<sub>3</sub> was estimated to vary from 0.8 in tropical rainforests to 0.5 in other forests to 0.2 for other vegetation. Riddick (2012) estimated the overall capture fraction at 59 % on soil and 73 % on vegetation from seabird-derived nitrogen experiments, taking account of different seabird habits. However, the capture of NH<sub>3</sub>-on vegetation is poorly constrained and is dependent to canopy features and boundary layer meteorology (Sutton et al., 2013). Because chicken manure is mostly applied to bare fields, there is not much vegetation capture of NH<sub>3</sub>-at the earlier stage, therefore this effect is not included in the present study. However, such an effect can be relevant for free range chicken that are kept outdoors under a woodland canopy (Bealey et al., 2014), so this effect would warrant further consideration if such practices became widespread.

Fourth, in addition to the atmospheric NH<sub>2</sub>-emission, canopy recapture of NH<sub>2</sub>-and the runoff, there are other processes influencing the nitrogen pathways, such as losses through nitrification and denitrification, that are not currently included in the AMCLIM-Poultry model. Nitrification is in general an aerobic process which is mainly influenced by the oxygen availability in the soils, with other controls on it including soil water content and soil temperature, while denitrification is generally an anaerobic process, dependent on soil porosity, soil water content, temperature and some other empirical coefficients (Butterbach Bahl et al., 2011). As the major objective of this study is to quantify the NH<sub>3</sub> emissions from practice relevant to chicken farming, these pathways have not been included.

## 4.45. Potential to consider NH3 mitigation scenarios.

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The process-based approach of the AMCLIM-Poultry model lends itself well to the opportunity to assess the implementation of possible management options to abate NH<sub>3</sub> emissions. Of the many measures for reducing NH<sub>3</sub> emissions as described by the UNECE (Bittman et al., 2014) several of them could be incorporated as part of future model development, e.g.:

- a) Measures to optimize animal diets, reducing excretion per animal. Such measures could be incorporated in the estimated amount of excretion per bird.
- b) Measures to reduce moisture in poultry houses, to reduce UA hydrolysis. Such measures could be incorporated into the relationship between indoor and outdoor conditions for relative humidity.
- c) Measures to reduce temperature of stored manure, to reduce UA hydrolysis and NH<sub>3</sub> emission. Such measures could be included in a possible future AMCLIM module on manure storage, by altering model temperature.
- d) Measures to alter the timing of manure application to favour land application under cool conditions. This could be included by altering assumed ambient temperature compared with seasonal averages.
- 30 e) Measures to incorporate poultry manure immediately into the soil. This could be included empirically based on alteration of atmospheric transfer resistances, or by more detailed development of several vertical layers or the model nitrogen pools (cf. Riedo et al., 2002).

While such considerations represent opportunities for future work, they highlight how a the AMCLIM-Poultry model is well suited to consideration of NH<sub>3</sub> emissions abatement scenarios.

#### **5 Conclusions**

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This paper presented the simulated NH<sub>3</sub> emission from global chicken farming by using the AMCLIM-Poultry model, including consideration of meteorological effects and simplified agricultural practices. The AMCLIM-Poultry model was designed based on underlying physics and chemistry, supported by evidence from experimental studies.

The magnitude of total NH<sub>3</sub> emissions from chicken farming estimated by the AMCLIM-Poultry based on 2010 was 5482.35.5 ± 1.2 Gg-Tg N yr<sup>-1</sup>, which accounts for approximately 13 ± 3 % of agriculture-derived NH<sub>3</sub> emissions (Crippa et al., 2016). High NH<sub>3</sub> emissions were from South and East Asia, Europe and southeast US. These regions also had high NH<sub>3</sub> volatilization rates, expressed as the percentage of excreted nitrogen (Pv) that is volatilized as NH<sub>3</sub>. The tropics often had high Pv values being up to five times than cold or dry regions, which illustrates how large NH<sub>3</sub> emission potentials are expected under hot and wet conditions. Agricultural activities related to chicken represent appreciable NH<sub>3</sub> sources, indicating that currently increasing NH<sub>3</sub> emissions accompanied by increasing chicken density (FAO, 2018) is important, especially as climate change is also expected to increase NH<sub>3</sub> emissions, as demonstrated by the spatial comparisons of the model.

Based on 2010, the model estimated that 24.222 % of the total excreted nitrogen was volatilized as NH<sub>3</sub> emission from chicken housing. The total NH<sub>3</sub> emission was 2185.5 G<sub>2.0</sub> Tg N, where 1374.71.3 Gg Tg N was from broilers and 810.0.78 TGg N was from layers. For the land based emissions, global NH<sub>3</sub> emissions were 2582.3 G<sub>2.7</sub> Tg N from manure fertiliser applications for crops and 714.50.7 TGg N from backyard chicken excretion, respectively, with strong spatial and temporal variations. In the current model approach, NH<sub>3</sub> emissions from manure storage are incorporated as 'in-house' storage with housing emissions. Further information on variation in practices is needed as a basis to estimate NH<sub>3</sub> emission from out-door storage of chicken manure, although the overall climate effect is expected to be midway between that for housing (covered outdoor storage) and land-spreading (uncovered storage).

Contrary to empirical approaches, this study uses a process-based method to quantify NH<sub>3</sub> emission from chicken, which provides a foundation for estimating emissions from other livestock types, based on theoretical considerations. The calculation of P<sub>V</sub> values is an asset of the model, which provides an insight of how environmental interactions will affect the NH<sub>3</sub> emissions, and which could also be applied to consider scenarios using emission abatement options. Strong spatial variation of P<sub>V</sub> implies that a single empirically derived emission factor would not usually reflect reality under different climate conditions. The results of this study show increased emissions under warm conditions, pointing to an expectation that climate change will increase chicken NH<sub>3</sub> emissions globally. The different relationships for housed chicken (primarily temperature and humidity dependence) and for backyard birds and manure spreading (primarily temperature and precipitation dependence), indicate that the net effect of climate change on regional emissions will depend on the relative composition of chicken types and management.

#### Data availability

Model results presented in this study are in netCDF format and can be freely accessed—(embargoed) through Edinburgh DataShare (<a href="https://datashare.is.ed.ac.uk/handle/10283/3644">https://datashare.is.ed.ac.uk/handle/10283/3774</a>, Jiang et al., 2020).

#### 5 Author contribution

JJ, DS and MAS designed the research. JJ developed the model code and performed the simulations. AU and GT prepared the model input data. JJ, DS and MS analysed the model outputs and wrote the paper. All authors contributed to the interpretation of results and critical revision.

## **Competing interest**

0 The authors declare they have no conflict of interest.

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# Appendix

Abbreviation	Unit	Model Variable
$f_N$	g N g excretion <sup>-1</sup>	N content of chicken excretion
$f_{\mathit{UA}}$		Fraction of uric acid in chicken excretion
$F_{e}$	$g N m^{-2} s^{-1}$	Total nitrogen excretion rate from chicken
$F_{H2O}$ (evap)	$g m^{-2} s^{-1}$	Evapouration
$F_{H2O}$ (rain)	$g m^{-2} s^{-1}$	Precipitation
$F_{NH3}$	$g N m^{-2} s^{-1}$	Net rate of conversion of TAN to gaseous NH <sub>3</sub> within litter/manure
$F_{removal}$	g N s <sup>-1</sup>	Removal of NH <sub>3</sub> through ventilation in the chicken house
$F_{surface}$	g N s <sup>-1</sup>	Total flux of NH <sub>3</sub> from surface litter in the chicken house
$F_{TAN}$	g N m <sup>-2</sup> s <sup>-1</sup>	Conversion rate of uric acid to TAN
K(T,pH,RH)	s <sup>-1</sup>	Function of temperauture, pH and RH influencing uric acid hydrolysis rate
$k_{pH}$		Function of pH influencing uric acid hydrolysis rate
$k_{RH}$		Function of RH influencing uric acid hydrolysis rate
$k_T$		Function of temperauture influencing uric acid hydrolysis rate
$m_E$		Equilibrium moisture content of litter/manure
$M_{\it available\ water}$	g m <sup>-2</sup>	Mass of water in the system that is available for washoff
$M_{excretion}$	g m <sup>-2</sup>	Mass of excretion
$M_{H2O}$	g m <sup>-2</sup>	Mass of water in the system
$M_{H2O}$ (e)	g m <sup>-2</sup>	Mass of water in the excretion
$M_N$	g N m <sup>-2</sup>	Mass of nitrogen components
$M_{N ext{-}runoff}$	g N m <sup>-2</sup>	Mass of instant runoff for nitrogen components
$M_{TAN}$	g N m <sup>-2</sup>	Mass of nitrogen in form of TAN
$M_{\mathit{UA}}$	g N m <sup>-2</sup>	Mass of nitrogen in form of uric acid
$N_{\it Crop}$	g N m <sup>-2</sup>	Amount of total N application for individual crops
$N_{\mathit{Crop\_Chicken}}$	$g N m^{-2}$	Amount of chicken manure-N application for individual crops
$N_{\it Soil\_Chicken}$	$g N m^{-2}$	Amount of available chicken manure-N
$N_{\it Total\_manure}$	$g N m^{-2}$	Amount of total N application for all crops
pH		pH of litter/manure
Q	$m^3 s^{-1}$	Ventilation rate in chicken house
$\mathcal{Q}_{\mathit{available}}$ water	mm	Pools of water in the system that is available for washoff
$r_N$	$\mathrm{mm}^{-1}$	Washoff factor
$R_{runoff}$		Runoff coefficient
$R^*$	s m <sup>-1</sup>	Overall indoor resistance in chicken house
$R_a$	s m <sup>-1</sup>	Aerodynamic resistance
$R_b$	s m <sup>-1</sup>	Boundary layer resistance
RH	%	Relative humidity
S	$m^2$	Surface area of chicken house
T	K	Ground temperature
V	m <sup>3</sup>	Volume of chicken house
$V_{H2O}$	ml m <sup>-2</sup>	Volume of water in the manure
z	m	Reference height
Xin	g m <sup>-3</sup>	Air concentrantion of NH <sub>3</sub> in chicken house
$\chi_{out}$	g m <sup>-3</sup>	Air concentrantion of NH <sub>3</sub> of embient environment
$\chi_{surface}$	g m <sup>-3</sup>	Concentrantion of NH <sub>3</sub> in litter/manure on the surface

Abbreviation	Unit	Model Variable
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$F_{H2O}$ (rain)	g m <sup>-2</sup> s <sup>-1</sup>	Precipitation
$F_{NH3}$	g N m <sup>-2</sup> s <sup>-1</sup>	Net rate of conversion of TAN to gaseous NH3 within litter/manure
$F_{removal}$	g N s <sup>-1</sup>	Removal of NH <sub>3</sub> through ventilation in the chicken house
$F_{surface}$	g N s <sup>-1</sup>	Total flux of NH <sub>3</sub> from surface litter in the chicken house
$F_{TAN}$	g N m <sup>-2</sup> s <sup>-1</sup>	Flux of TAN from uric acid hydrolysis
K(T,pH,RH)	$s^{-1}$	Function of temperauture, pH and RH influencing uric acid hydrolysis rate
$k_{pH}$		Function of pH influencing uric acid hydrolysis rate
$k_{RH}$		Function of RH influencing uric acid hydrolysis rate
$k_T$		Function of temperauture influencing uric acid hydrolysis rate
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$M_{excretion}$	g m <sup>-2</sup>	Mass of excretion
$M_{H2O}$	g m <sup>-2</sup>	Mass of water in the system
$M_{H2O}$ (e)	g m <sup>-2</sup>	Mass of water in the excretion
$M_N$	g N m <sup>-2</sup>	Mass of nitrogen components
$M_{N ext{-runoff}}$	g N m <sup>-2</sup>	Mass of instant runoff for nitrogen components
$M_{TAN}$	g N m <sup>-2</sup>	Mass of nitrogen in form of TAN
$M_{UA}$	g N m <sup>-2</sup>	Mass of nitrogen in form of uric acid
$N_{Crop}$	g N m <sup>-2</sup>	Amount of total N application for individual crops
$N_{Crop\_Chicken}$	g N m <sup>-2</sup>	Amount of chicken manure-N application for individual crops
$N_{Available}$	g N m <sup>-2</sup>	Amount of available chicken manure-N
$N_{Total\_manure}$	g N m <sup>-2</sup>	Amount of total N application for all crops
pH	8	pH of litter/manure
Q	$m^{3} s^{-1}$	Ventilation rate in chicken house
${\cal Q}_{available\ water}$	mm	Pools of water in the system that is available for washoff
$r_N$	mm <sup>-1</sup>	Washoff factor
$R_{runoff}$		Runoff coefficient
R*	s m <sup>-1</sup>	Overall indoor resistance in chicken house
$R_a$	s m <sup>-1</sup>	Aerodynamic resistance
$R_b$	s m <sup>-1</sup>	Boundary layer resistance
RH	%	Relative humidity
S	$m^2$	Surface area of chicken house
T	K	Ground temperature
V	$m^3$	Volume of chicken house
$V_{H2O}$	ml m <sup>-2</sup>	Volume of water in the manure
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$\chi_{in}$	g m <sup>-3</sup>	Air concentrantion of NH <sub>3</sub> in chicken house
$\chi_{out}$	g m <sup>-3</sup>	Air concentrantion of NH <sub>3</sub> of embient environment
$\chi_{surface}$	g m <sup>-3</sup>	Concentrantion of NH <sub>3</sub> in litter/manure on the surface

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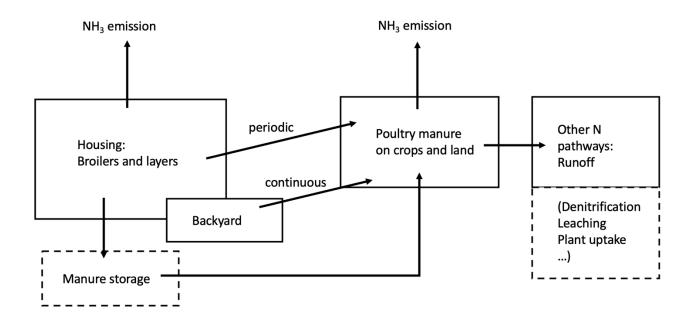


Figure 1 Schematic of the AMCLIM-Poultry model for estimating NH3 emissions from global chicken farming following nitrogen pathways from chicken farms to land spreading. <u>Arrows represent the nitrogen flows from chicken farming.</u> Aspects noted in dashed boxes are not investigated in this study.

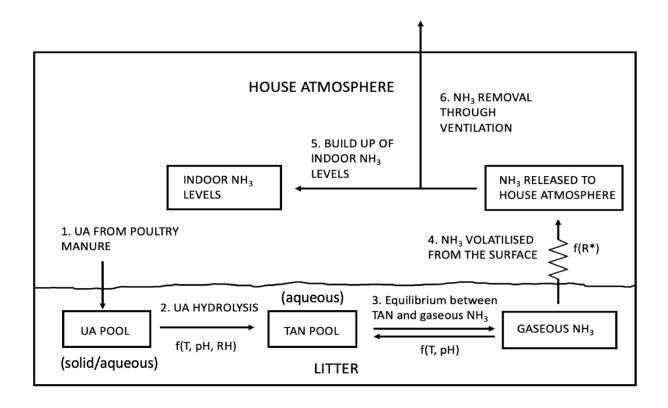


Figure 2 Schematic of  $NH_3$  volatilization in the poultry house. UA is uric acid; TAN is total ammoniacal nitrogen,  $R^*$  is the resistance for gaseous transfer from the litter surface to the in-house atmosphere (adapted from Elliott and Collins, 1982)

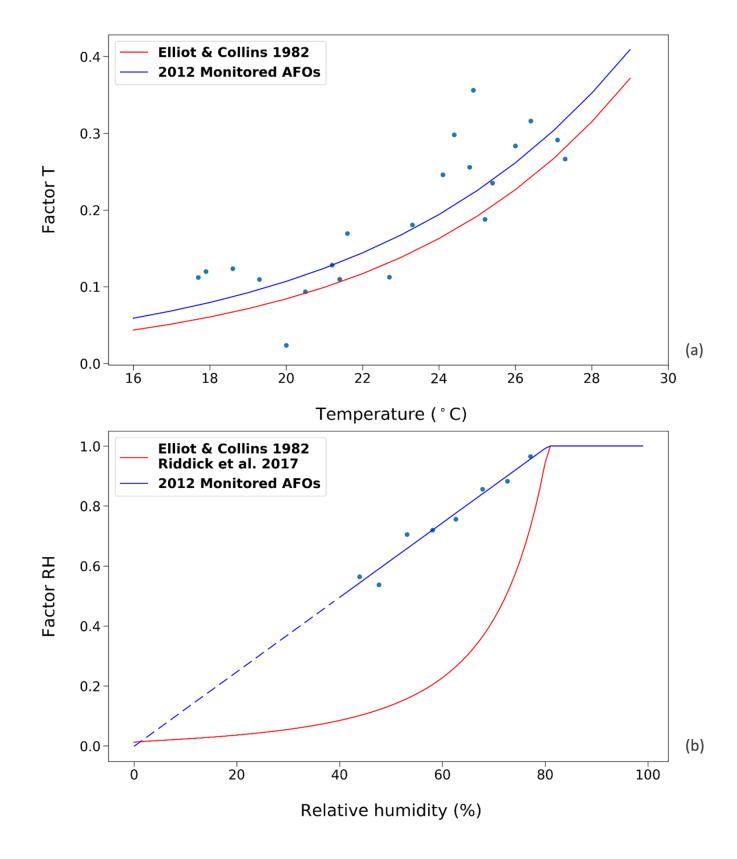
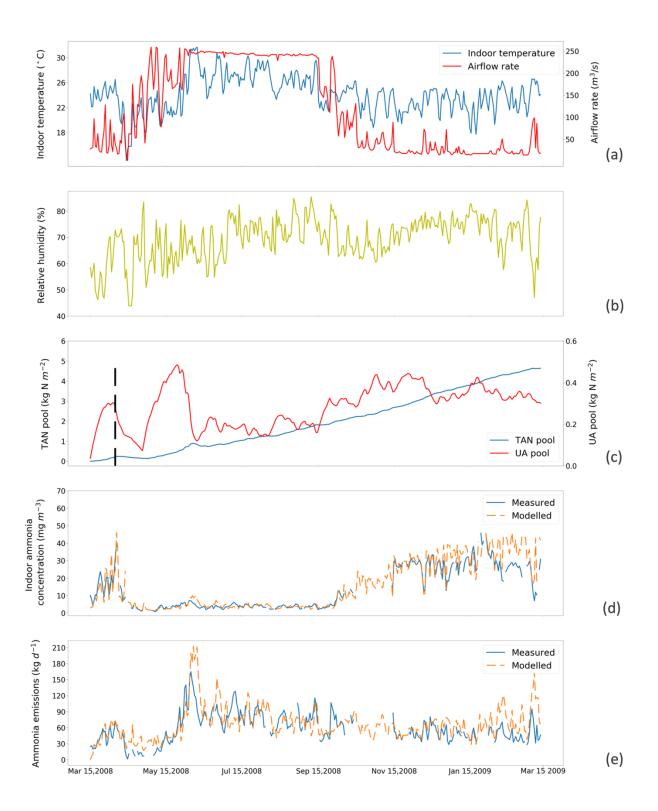


Figure 3 Factors affecting UA hydrolysis rate in chicken houses. Red curves represent the results from Elliott & Collins, 1982. Blue curves represent results from this study using data from the 2012 Monitored AFOs (see Sect. 2.2.1). a) Influence of temperature on UA hydrolysis. b) Influence of relative humidity on UA hydrolysis at optimum temperature condition (≥35 °C). Dashed line is the extrapolation of factor RH as a function of RH due to lack of data when relative humidity was below 40 % in the AFO experiments.



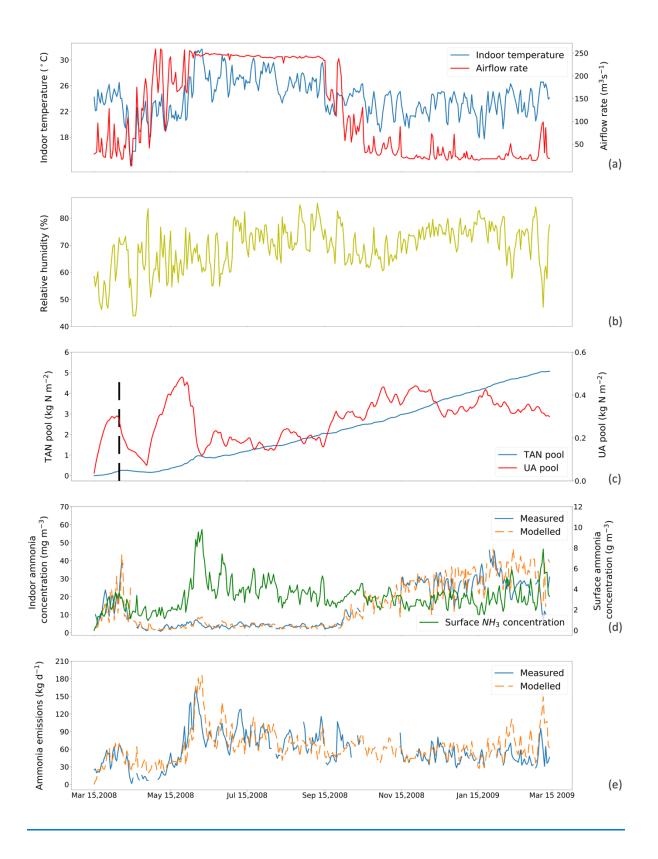


Figure 4 Site simulations using fixed resistance (R\*) value of 16700 s m<sup>-1</sup> for House A at site NC2B, Nash, North Carolina from March 15 to March 15, 2009. a) Measured daily mean indoor temperature and airflow rate of the house. b) Measured daily mean relative humidity of the house. c) Modelled TAN pool and UA pool. The black dashed line indicates the house emptying date of April/09/2008. d) Comparison between measured and modelled indoor NH<sub>3</sub> concentrations of the house, and surface NH<sub>3</sub> concentrations. e) Comparison between modelled NH<sub>3</sub> emissions and calculated NH<sub>3</sub> emissions from measured indoor concentrations. The simulation illustrated uses the new parametrization (based on the AFO data, Fig. 3) for relative humidity dependence of UA hydrolysis.

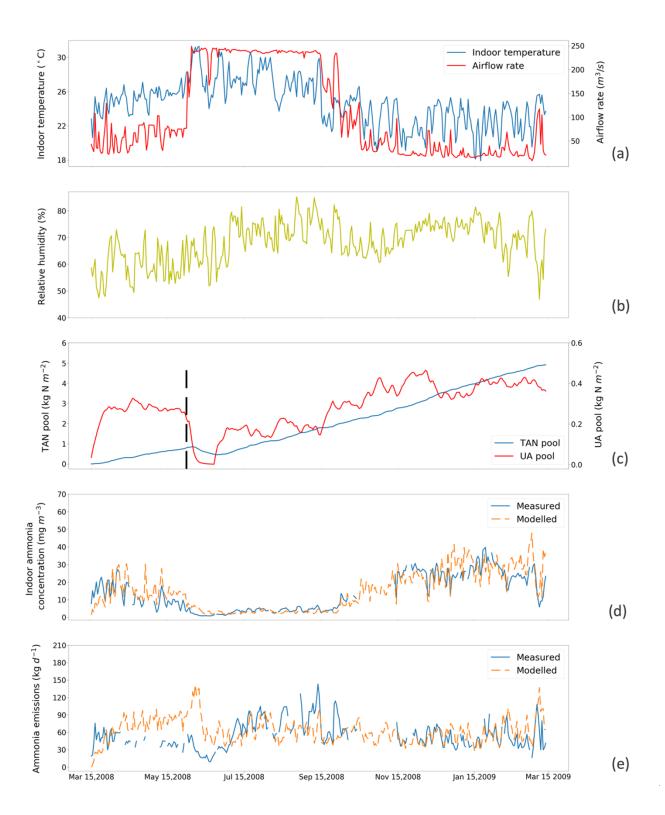




Figure 5 The same as Fig. 4, but for Site-simulations using fixed resistance (R\*) value of 14369 s m<sup>-1</sup> for House B at site NC2B, Nash, North Carolina from March 15 to March 15, 2009. a) Measured daily mean indoor temperature and airflow rate of the house. b) Measured daily mean relative humidity of the house. c) Modelled TAN pool and UA pool. The black dashed line indicates the house emptying date of June/03/2008. d) Comparison between measured and modelled indoor NH<sub>3</sub> concentrations of the house. c) Comparison between modelled NH<sub>3</sub>-emissions and calculated NH<sub>3</sub>-emissions from measured indoor concentrations. The simulation illustrated uses the new parametrization (based on the AFO data, Fig. 3) for relative humidity dependence of UA hydrolysis.

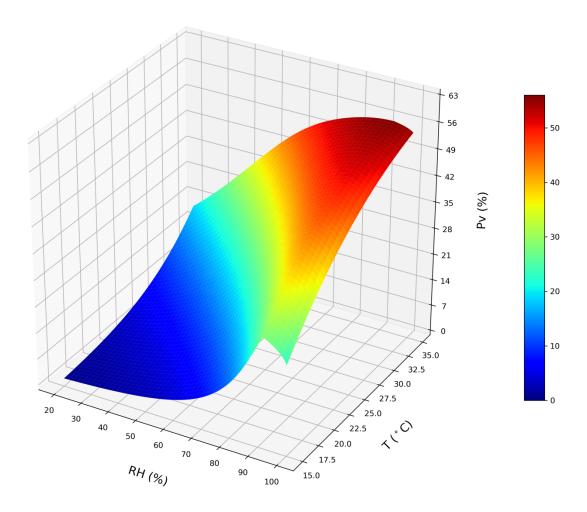


Figure 6 A conceptual 3-D sketch of  $NH_3$  volatilization rate  $(P_V(\%))$  that is driven by temperature (T) and relative humidity (RH) The surface plot is derived from a set of idealised steady state simulations with zero precipitation to simulate dependences for emissions from chicken housing (see Sect. 3.1.22.2.1 Shown using the new parametrizations for T and RH).

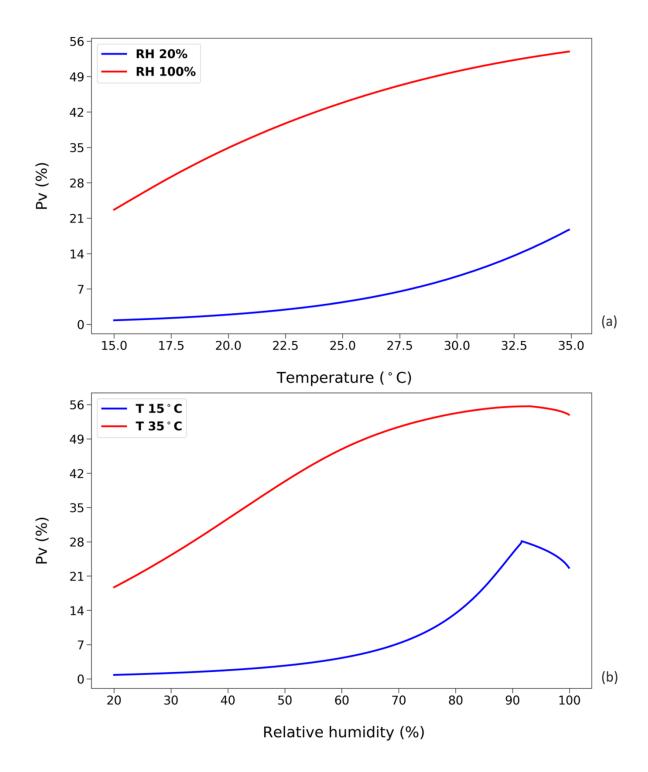


Figure 7 Curves that represent NH<sub>3</sub> volatilization rate ( $P_V$ , %) for 4 different temperature and RH regimes from Fig. 6based on annual idealised simulations (see Fig. 6). a) The NH<sub>3</sub> volatilization rate ( $P_V$  (%)) under dry (20 % relative humidity, RH) and wet (100 % RH) conditions, respectively. b) The NH<sub>3</sub> volatilization rate ( $P_V$  (%)) under 15 °C and 35 °C, respectively. (See Sect. 3.1.22.2.1, shown using the new parametrizations for temperature and RH).

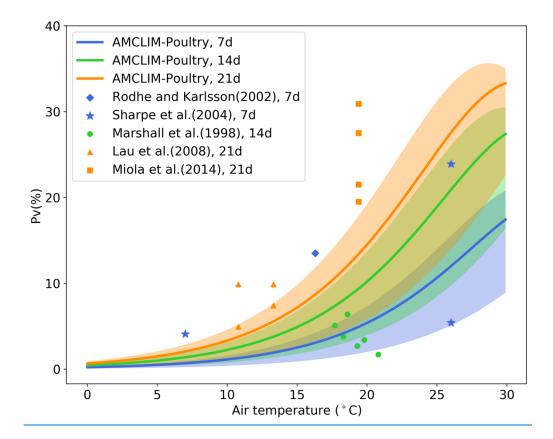
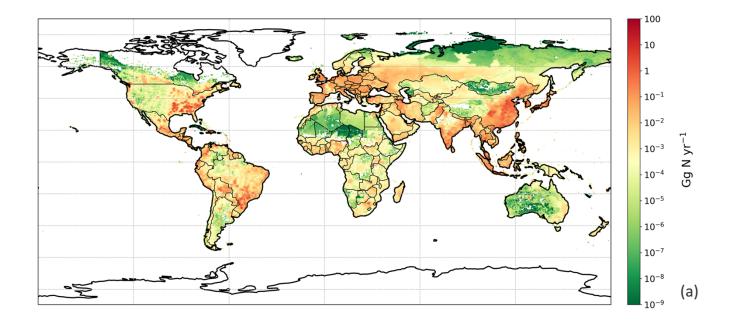
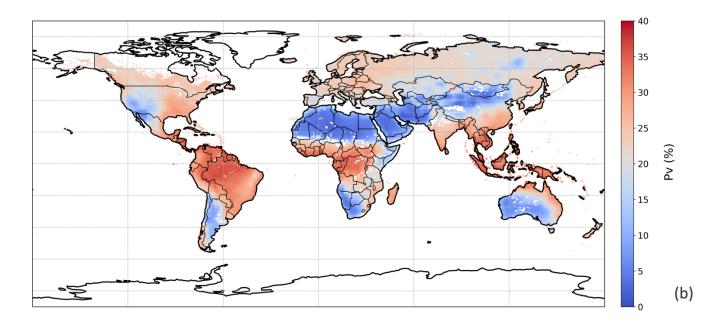


Figure 8 Simulated fraction of total applied nitrogen that is loss as NH<sub>3</sub>-N (PV, %) as a function of air temperature (°C) by the AMCLIM-Poultry for simulating periods of 7, 14 and 21 days, and comparison with experimental studies that measured NH<sub>3</sub>-N loss for 7, 14 and 21 days. Simulations conducted for rain-free conditions, where shaded areas indicate the range for simulations from 20 % to 100% relative humidity. The measured figure of 5 % volatilization at 27 °C by Sharpe et al. (2004) was associated with high precipitation not representative of these simulations.





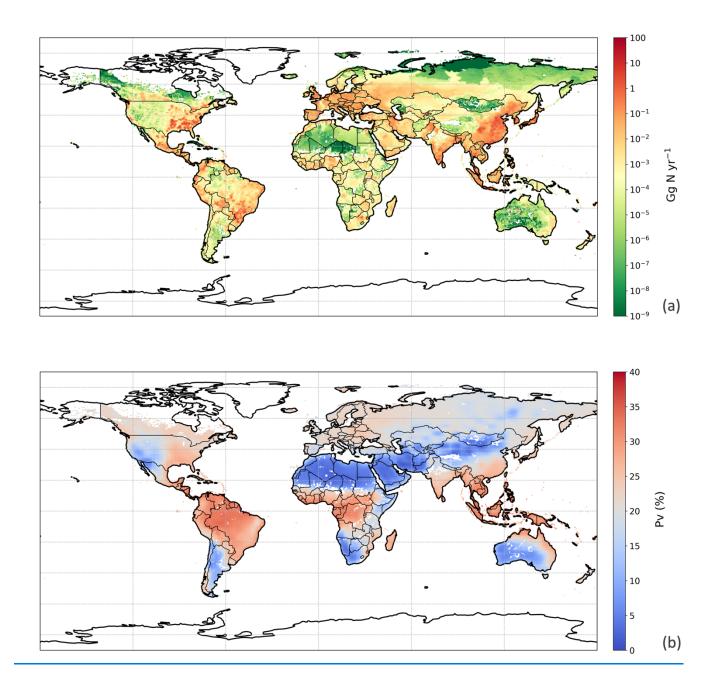
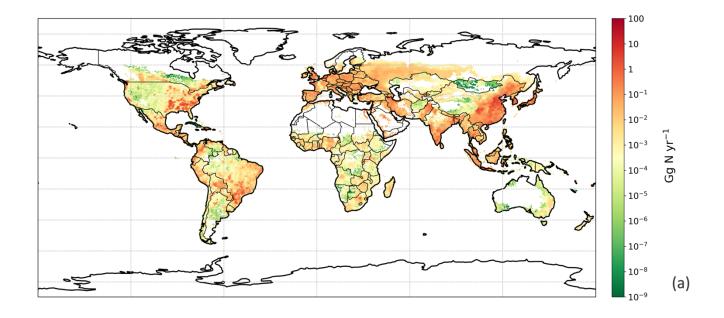
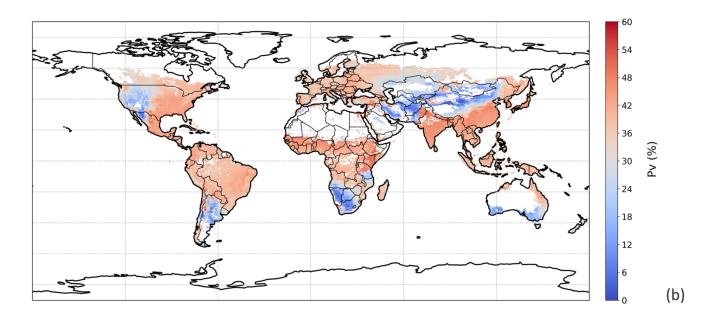


Figure 8-9\_Simulated a) annual global NH<sub>3</sub> emissions (Gg N yr<sup>-1</sup>) from chicken housing in 2010. b) Percentage of excreted nitrogen that volatilizes ( $P_V$ , %) as NH<sub>3</sub> from chicken housing in 2010. The resolution is  $0.5^{\circ} \times 0.5^{\circ}$ . For the simulation shown the RH parametrization for UA hydrolysis is taken from Elliott and Collins (1984). Figure S9 shows the results of using the RH parametrization based on new parameterization from AFOs monitored data, for comparison.





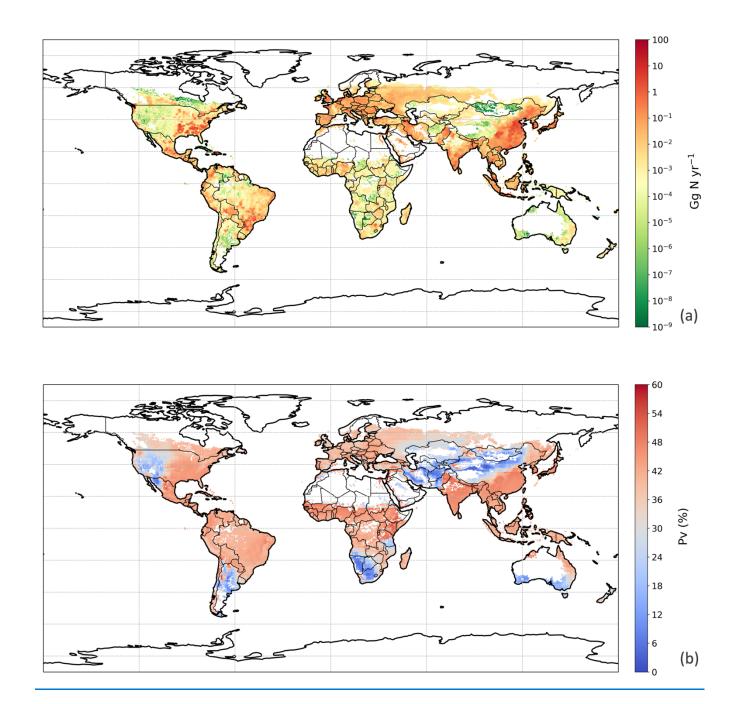
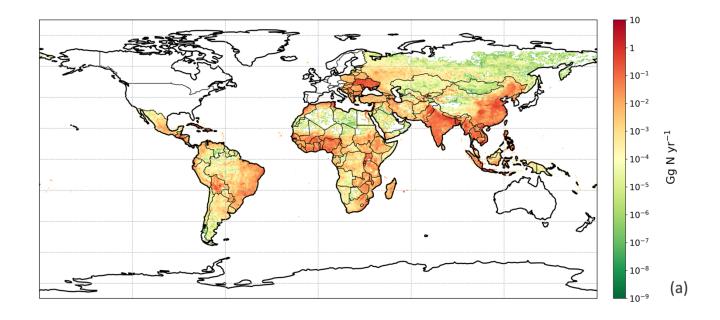
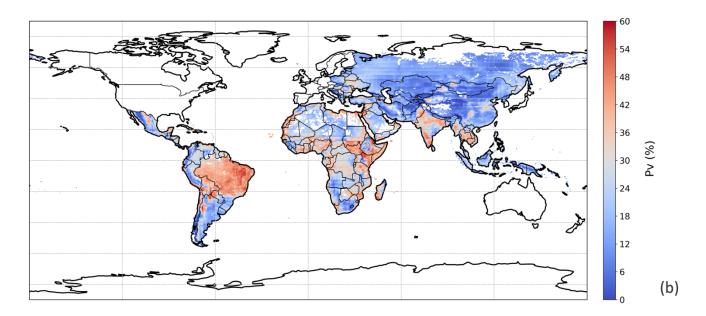


Figure 9-10 Same as Fig. 9, but for Simulated a) annual global  $NH_3$  emissions (Gg N yr $^4$ ) from chicken manure application for crops in 2010. b) Percentage of exercted nitrogen that volatilizes ( $P_V$ , %) as  $NH_3$  from chicken manure application for crops in 2010. The resolution is 0.5°×0.5°.





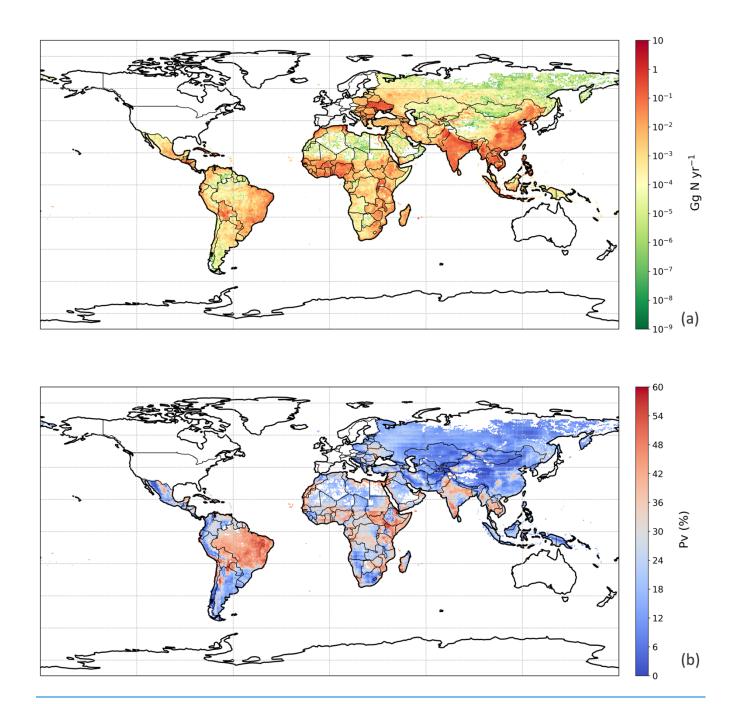
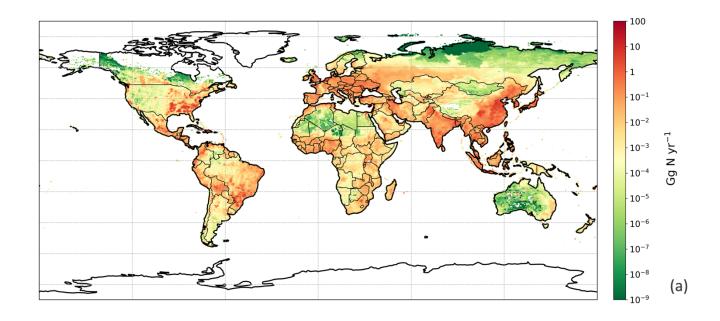
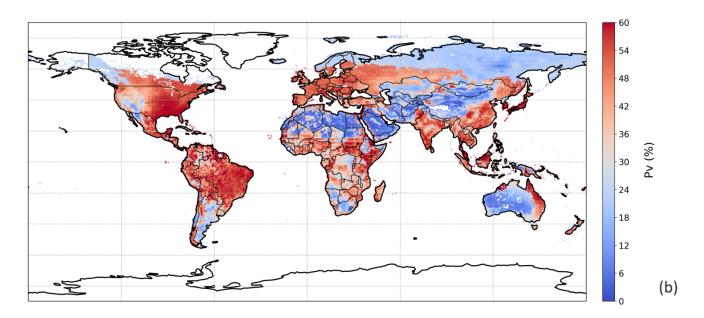


Figure 10-11 Same as Fig. 9, but for Simulated a) annual global NH<sub>3</sub>-emissions (Gg N yr<sup>-1</sup>) from-backyard chicken in 2010. b) Percentage of exercted nitrogen that volatilizes ( $P_V$ , %) as NH<sub>3</sub> from backyard chicken in 2010. The resolution is 0.5°×0.5°.





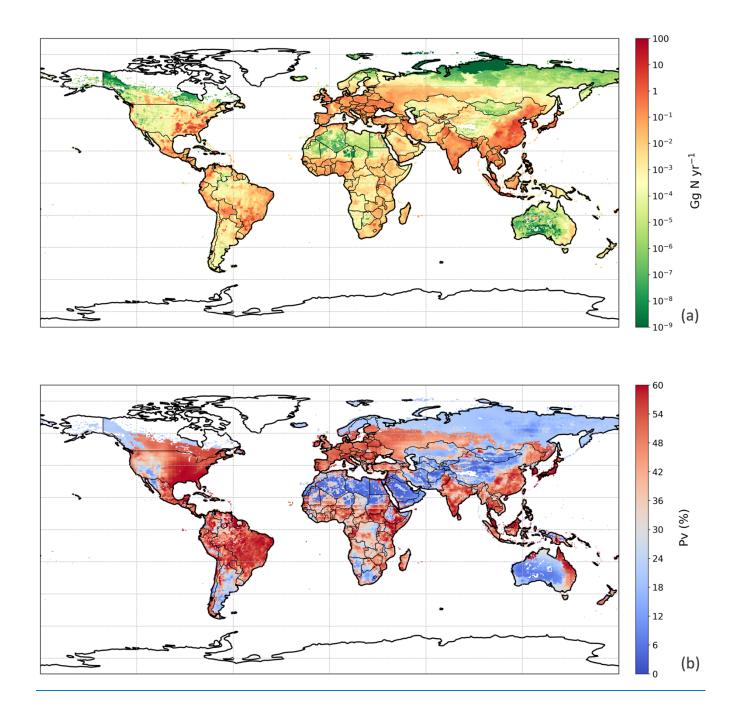
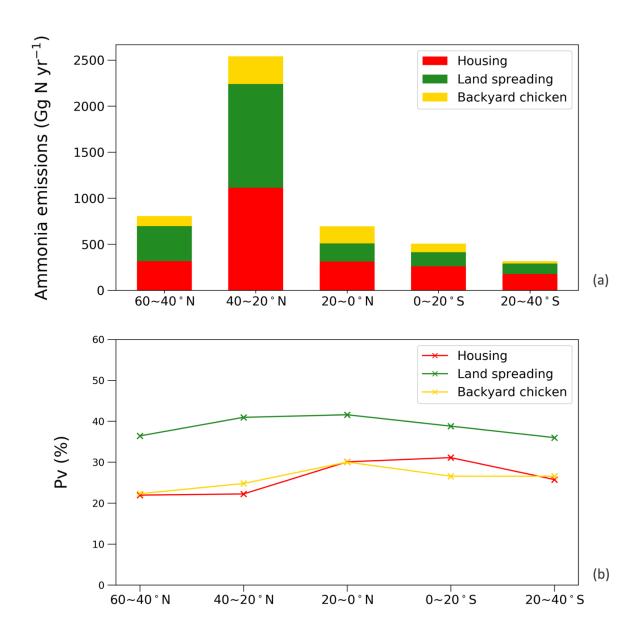


Figure  $\frac{11-12}{2}$  Simulated a) annual global NH<sub>3</sub> emissions (Gg N yr<sup>-1</sup>) from chicken agriculture in 2010. b) Percentage of excreted nitrogen that volatilizes (P<sub>V</sub>, %) as NH<sub>3</sub> from chicken agriculture in 2010. The resolution is  $0.5^{\circ}\times0.5^{\circ}$ .



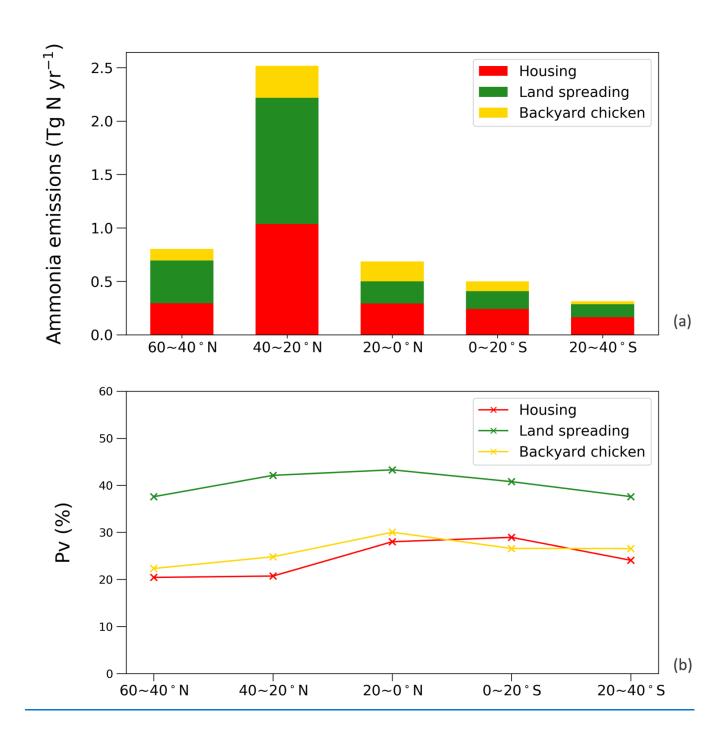
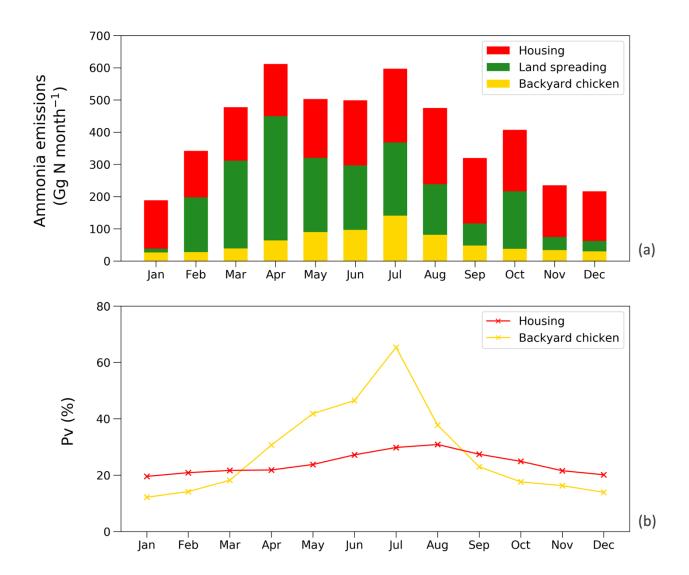


Figure  $\frac{12}{13}$  Simulations for chicken housing, manure applications to crops and land spreading of backyard chicken manure in 2010 given in regions. a) annual global NH<sub>3</sub> emissions ( $\frac{\text{Gg Tg}}{\text{Tg}}$ N yr<sup>-1</sup>). b) Percentage of excreted nitrogen that volatilizes (P<sub>V</sub>, %) as NH<sub>3</sub>.



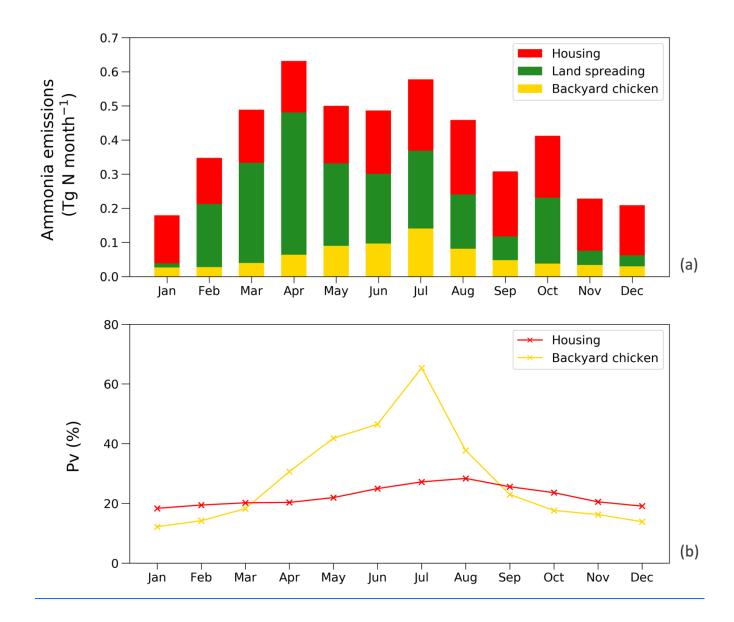


Figure  $\frac{13-14}{4}$  a) Monthly NH<sub>3</sub> emissions ( $\frac{\text{Gg Tg}}{\text{Ig}}$ N yr<sup>-1</sup>) from chicken housing, manure applications to crops and land spreading of backyard chicken manure in 2010. b) Percentage of excreted nitrogen that volatilizes ( $P_V$ , %) as NH<sub>3</sub> monthly for chicken housing and land spreading of backyard chicken manure.

Production system	Total excreted nitrogen (Tg N)	<u>Practice</u>	Total emission (Tg N)		Average P <sub>V</sub> (%)	
Broiler and layer	9.0 [±0.9]	Housing	2.0 [±0.6]		22 [±7] %	
		Land spreading	2.7 [±0.5]		39 [±7]* %	
Backyard chicken	2.2 [±0.2]	Left on land		0.7 [±0.2]	32 [±7]	<u>%</u>
<u>Total</u>	11.2 [±1.1]		5.5 [±1.2]		49 [±11	] %
Production system	Excreted nitroger (Gg N)	Practice		Emission (Gg N)		
Broiler and layer	9017.1	Housing	Housing		2185.5	
	<del>7017.1</del>	Land spreadi	Land spreading		<del>2582.3</del>	
Backyard chicken	<del>2178.3</del>	Left on lan	<del>d</del>	714.5		
<del>Total</del>	11195.4			5482.3	}	

Table 2 Estimates of NH<sub>3</sub> emissions from poultry/chicken farming by IDA for Denmark (Albrektsen et al., 2017) and by NARSES (Misselbrook et al., 2011) for the United Kingdom based on 2010, and by NEMA (Velthof et al., 2012) for Netherlands based on 2009\*. Ranges given in the P<sub>V</sub>-housing represents the geographical variations across the country.

Country	Ammonia emission from Housing (Gg N yr-1)	Ammonia emission from Spreading (Gg N yr <sup>-1</sup> )	Total excreted N (Gg N yr <sup>-1</sup> )	P <sub>V</sub> -housing (%)
Denmark	3.0 (IDA)	Not available	11.3 (IDA)	<u>26.5</u>
Denmark	<u>1.7 (AMCLIM)</u>	2.4 (AMCLIM)	7.9 (GLEAM)	21.5 (20.4 – 22.9)
Netherlands	11.4* (NEMA)	1.8* (NEMA)	62.9* (NEMA)	<u>18.1*</u>
	10.0 (AMCLIM)	15.0 (AMCLIM)	49.0 (GLEAM)	20.4 (20.0 – 21.0)
<u>United</u>	15.0 (NARSES)	<u>14.7 (NARSES)</u>	Not available	<u>17.8</u>
Kingdom	<u>17.4 (AMCLIM)</u>	23.7 (AMCLIM)	84.1 (GLEAM)	20.7 (18.6 – 22.1)

Table 3 Sensitivity test for model parameters for global application of the model.

<u>Parameter</u>	Value tested	Value change	$\Delta NH_3$ emission %	
	16700 s m <sup>-1</sup> (base)	<u>1 x</u>	0.0 %	
a, b Indoor resistance, R*	8350 s m <sup>-1</sup>	<u>0.5 x</u>	<sup>a</sup> 27.1 % a, b 8.5 %	
	33400 s m <sup>-1</sup>	<u>2 x</u>	a -30.6 % a, b -6.4 %	
a, b, c Manure pH (H <sup>+</sup> )	8.5 (base)	<u>1 x</u>	0.0 %	
pri (ii )	<u>7.5</u>	<u>0.1 x</u>	<u>-15.9 %</u>	

	9.5	<u>10 x</u>	<u>5.8 %</u>
b, c Runoff coefficient,	1 % mm <sup>-1</sup> (base)	<u>1 x</u>	0.0 %
R <sub>runoff</sub>	0.5 % mm <sup>-1</sup>	<u>0.5 x</u>	<u>16.5 %</u>
<u> 1Stunori</u>	2 % mm <sup>-1</sup>	<u>2 x</u>	<u>-11.8 %</u>
	11.2 Tg N year-1 (base)	<u>1 x</u>	0.0 %
a, b, c Excreted nitrogen	10.1 Tg N year <sup>-1</sup>	<u>0.9 x</u>	<u>-12.3 %</u>
	12.3 Tg N year <sup>-1</sup>	<u>1.1 x</u>	<u>12.6 %</u>

<sup>&</sup>lt;sup>a</sup> Parameters affect NH<sub>3</sub> emissions from housing. <sup>b</sup> Parameters affect NH<sub>3</sub> emissions from land spreading of chicken manure. <sup>c</sup> Parameters affect NH<sub>3</sub> emissions from backyard chicken.