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# Supplement of

# A dynamical process-based model for quantifying global agricultural ammonia emissions — AMmonia—CLIMate v1.0 (AMCLIM v1.0) — Part 2: Livestock farming

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# S1 Two-film model for the gas exchange across the air-liquid interface

The two-film model proposed by Liss and Slater (Liss and Slater, 1974) for estimating the gaseous flux across the air-liquid interface is used to model the NH<sub>3</sub> emissions from pit storage in animal houses and lagoon systems in the AMCLIM model because these systems hold large amount of water (as mentioned in Sections XXX and XXX). Figure S1 illustrates the flux of NH<sub>3</sub> is transferred from the liquid to the air across the interface. The main body of the liquid is assumed to be well-mixed, so the main resistances are from the gas and liquid phase interfacial layers (gas and liquid "films"). There are two transport processes. The first process is TAN from the bulk liquid to the interface ( $F_{\text{TAN to surface}}$ ) through molecular transfer that is driven by concentration gradients, which can be expressed as:

$$F_{\text{TAN to surface}} = k_{\text{L}}([\text{TAN (aq)}]_{\text{bulk liquid}} - [\text{TAN (aq)}]_{\text{interface}}),$$
 (S1)

where  $k_{\rm L}$  (m s<sup>-1</sup>) is an aqueous transfer coefficient for TAN (NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup>). The second process is NH<sub>3</sub> transported from the interface to the atmosphere (house atmosphere for housing simulations and free atmosphere for lagoon simulations), which can be expressed as:

$$F_{\text{NH}_3} = k_{\text{G}}([\text{NH}_3(g)]_{\text{interface}} - [\text{NH}_3(g)]_{\text{in/atm}}), \tag{S2}$$

where  $k_G$  (m s<sup>-1</sup>) is a gaseous transfer coefficient for NH<sub>3</sub>. The aqueous TAN concentration and the gaseous NH<sub>3</sub> concentration at the interface is in equilibrium as shown in Equation 3.3, and it is assumed that the transfer of NH<sub>3</sub> across the interface is in a steady state so that the two transport processes in aqueous and gaseous phase are equivalent.

$$k_{L}([TAN (aq)]_{bulk \, liquid} - [TAN (aq)]_{interface}) = k_{G}([NH_{3}(g)]_{interface} - [NH_{3}(g)]_{in/atm}). \tag{S3}$$

In AMCLIM, the calculations of NH<sub>3</sub> emissions and other transport processes such as diffusion use resistances, some of which are the reciprocals of the transfer coefficients as shown in Equation 4.3. In addition, as NH<sub>3</sub> emissions take place from wet surfaces, the gaseous NH<sub>3</sub> concentration at the interface in the two-film model is represented by  $\chi_{srf}$  in AMCLIM. By combining Equations A.49 to A.51, the NH<sub>3</sub> emission can be calculated by simulating the TAN concentration of the bulk liquid using the following equation (under the simplification that atmospheric indoor NH<sub>3</sub> concentration are 0; as expressed by Equation 4.23):

$$F_{\text{NH}_3} = \frac{\chi_{\text{srf}}}{R_{\text{G}}} = \frac{[\text{TAN (aq)}]}{R_{\text{GL}}},\tag{S4}$$

25 where  $R_{\rm GL}$  is a combined resistance that limits the NH<sub>3</sub> transfer across the gas-liquid interface, which is expressed as:

$$R_{\rm GL} = \frac{1}{k_{\rm L}} + \frac{1}{k_{\rm G}K_{\rm NH_2}}.$$
 (S5)

The aqueous and gaseous transfer coefficients are empirically derived (Ni, 1999), which are calculated by the following equations:

$$k_{\rm L} = 1.417 \times 10^{-12} T^4,$$
 (S6)

$$30 k_{\rm G} = 0.001 + 0.0462u_*Sc^{0.67}, (S7)$$

where Sc is the Schmidt number which is calculated from the kinematic viscosity ( $\nu$ , m<sup>2</sup> s<sup>-1</sup>) and diffusivity of NH<sub>3</sub> as follows:

$$Sc = \frac{v}{D_{\text{NH}_3}^{\text{gas}}},\tag{S8}$$

$$v = 1.56 \times 10^{-5} \left(\frac{T + 273.15}{298.15}\right)^{\frac{3}{2}}.$$
 (S9)

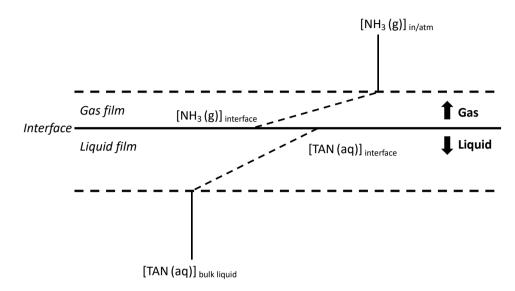


Figure S1. Sketch of the ammonia transfer processes across an air-liquid interface (adapted from Liss and Slater (1994)). In AMCLIM,  $[NH3(g)]_{interface}$  in the figure is represented by  $\chi_{srf}$ , and  $[NH3(g)]_{in/atm}$  is represented by  $\chi_{in}$  or  $\chi_{atm}$ .

#### S2 Hydrolysis of urea/uric acid and mineralization of organic nitrogen

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A general term,  $K_N(s^{-1})$  is used in Eq.(6) for expressing the conversion rates of multiple nitrogen forms to TAN, e.g., urea, UA and organic nitrogen. These processes are strongly dependent on the environmental factors, such as temperature, RH, water content and the pH of soils or manure. The hydrolysis rate of urea ( $K_{Urea}$ , s<sup>-1</sup>) is parameterized as follows by assuming a first order reaction according to (Sherlock and Goh, 1984):

$$\frac{\mathrm{d}M_{\mathrm{Urea}}}{\mathrm{d}t} = -K_{\mathrm{Urea}}M_{\mathrm{Urea}},\tag{S10}$$

$$K_{\text{Urea}} = 1 - \exp\left(-k_h \cdot WFPS \cdot A_h\right), \tag{S11}$$

$$A_h = 0.25 \exp(0.0693 (T - 273.15)),$$
 (S12)

where  $k_h$  is the urea hydrolysis constant for urine (6.4×10<sup>-5</sup> s<sup>-1</sup> or 0.23 h<sup>-1</sup>; Sherlock and Goh, (1984)) and for urea in soils (8.3×10<sup>-6</sup> s<sup>-1</sup> or 0.03 h<sup>-1</sup>; Dutta et al. (2016)). Real urine from animals is found to have a faster decomposition rate than chemical urea fertilizer (Haynes and Williams, 1993; Sherlock and Goh, 1985). *WFPS* is the water-filled pore space and is set to 1 for livestock urine.  $A_h$  is a temperature correction dependence, and T is the temperature in Kelvin (K).

The hydrolysis rate of uric acid ( $K_{UA}$ , s<sup>-1</sup>) is calculated from the product of a series of conversion rate functions (Elliott and Collins, 1982), as follows:

$$K_{\text{IIA}} = 0.2k_{nH}k_Tk_{RH},$$
 (S13)

where  $k_{pH}$ ,  $k_T$  and  $k_{RH}$  are the functions of pH, temperature and RH influencing uric acid hydrolysis rate, respectively. The maximum estimated hydrolysis rate of uric acid is 0.2 d<sup>-1</sup>. The temperature (in °C), RH and pH dependence of UA hydrolysis rate is shown by the following equations:

$$k_T = \frac{exp^{(0.149(T-273.15)+0.49)}}{exp^{(0.149(35)+0.49)}}$$
(S14)

The temperature dependence follows an exponential relationship and is normalised to the maximum rate at 35 °C (Jiang et al., 2021).

$$60 \quad k_{RH} = 0.0124 \, RH - 0.0014 \tag{S15}$$

The RH dependence increases linearly as RH increases, reaching the maximum rate of 1 at RH 80 % (Jiang et al., 2021). Note that the humidity level can be a key limiting factor in determining the rate of uric acid hydrolysis and subsequent TAN emissions.

$$k_{pH} = \frac{1.34(\text{pH}) - 7.2}{1.34(\text{g}) - 7.2} \tag{S16}$$

65 A fixed pH of 8.5 is the typical value of poultry manure (Elliott and Collins, 1982; Sommer and Hutchings, 2001).

Organic N is categorised into three types: a) available organic nitrogen, b) resistant organic nitrogen and c) unavailable organic nitrogen, referring to how readily that the organic nitrogen is available to decompose to form TAN (Riddick et al., 2016). A fraction of 50 % organic nitrogen is assumed to be available organic nitrogen, 45 % is in the resistant form, and the rest of 5 % goes to the unavailable nitrogen pool (Riddick et al., 2016). The rate of mineralization of organic nitrogen is determined by the following equation:

$$K_{\text{OrgN}} = B_{a,r} A_m, \tag{S17}$$

$$A_m = t_{r1} \exp(t_{r2}(T - 273.15)), \tag{S18}$$

where  $B_{\rm a,r}$  ( $B_{\rm a} = 8.94 \times 10^{-7} \, {\rm s}^{-1}$ ;  $B_{\rm r} = 6.38 \times 10^{-8} \, {\rm s}^{-1}$ ) are the mineralization constants for available and resistant organic N (Gilmour et al., 2003; Vigil and Kissel, 1995).  $A_{\rm m}$  is a temperature correction dependence, with  $t_{\rm r1}$  and  $t_{\rm r2}$  are equivalent to 0.0106 K<sup>-1</sup> and 0.12979 K<sup>-1</sup>, respectively.

# S3 Housing simulations for pigs, poultry and ruminants

# S3.1 Simulations for pig housing

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AMCLIM-Housing includes three types of animal houses, as introduced in Section 2.2.1. The first two housing types are designed for pig housing simulations. The first housing type has slatted floor and pit storage that allows pig excreta to be stored in-situ, keeping the floor area clean. For this housing type, a two-source emission scheme is used to model NH<sub>3</sub>

emissions as there are two emitting surfaces: the slats and pit. The two NH<sub>3</sub> emission elements are treated as additive, i.e., the total housing emission is the sum of the emissions from the two housing compartments. To represent the processes on the slats and in the pit, the pools of N species and other simulated variables are divided into two separate reservoirs. Pig excreta are split proportionally between the two reservoirs depending on the gap space of the slats. For example, if the gap space is 20 %, then 20 % of initial pig excreta will fall into the underneath pit, and the remaining 80 % will stay on the slats. Given the fact that excretions left on the slats will eventually fall to the pit (i.e., through cleaning), but excretions in the pit cannot go back to the slatted floor above, a uni-directional transfer is applied on a daily basis in AMCLIM–Housing. It is assumed that all pools from the slat reservoir go into pit reservoir by the end of each day, and the slat reservoir is reset to zero subsequently. Excreta that go into the pit are stored for longer time, e.g., weeks to months.

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The process of NH<sub>3</sub> volatilization differs between the two reservoirs because of the different amount of water held in the two reservoirs. For the slats, excreta are typically a thin wet layer, so the surface concentrations can be expressed by the concentrations of the entire layer. The gaseous NH<sub>3</sub> concentration at the surface is directly derived from the aqueous TAN concentration of this layer. In contrast, the pit reservoir holds more water (and faeces) because urine in the excreta accumulates in the pit. There is an additional aqueous transfer process of TAN from the bulk water to the air-water interface.

AMCLIM-Housing incorporates a two-film model that describes the gas exchange across the air-liquid interface (Liss, 1973; Liss and Slater, 1974). Details of the two-film model and the calculation of mass transfer are given in Appendix A10.

The second house type is a normal barn with a solid floor (without pit storage). In AMCLIM-Housing, normal barns are assumed to be cleaned daily so that pig excreta are removed from the house, and all pools are reset to zero every day. Volatilization of NH<sub>3</sub> from the pig excreta on the solid floor is identical to the processes taking place on the slats in the first house type.

For pigs (and ruminants), the water pool simulated in AMCLIM-Housing is determined by sources of water from urination ( $F_{\text{urine}}$ ), water in faecal excreta ( $F_{\text{faecal water}}$ ) and loss by evaporation of water ( $F_{\text{evap}}$ , mm s<sup>-1</sup>) A cleaning-day function is included in the equation to account for the effect of cleaning on the water pool as follows:

$$\frac{dM_{H_2O}}{dt} = F_{\text{urine}} + F_{\text{faecal water}} - F_{\text{evap}} - \psi_{\text{cleaning}}(t, H_2O)$$
 (S19)

105 Excess water, such as washing water or drinking water in the houses, is not included since the quantity is unknown.

The evaporation rate in the animal houses is approximated by applying an aerodynamic method using a vapor transfer coefficient ( $B_{\text{vap}}$ , m Pa<sup>-1</sup> s<sup>-1</sup>) and vapor pressure deficit as follows (Chow et al., 1988):

$$F_{\text{evap}} = B_{\text{vap}}(e_{\text{s}} - e_{\text{a}}), \tag{S20}$$

$$B_{\text{vap}} = \frac{0.622k^2 \rho_{\text{air}} u}{\rho_{\text{water}} p[\ln{(\frac{Z}{Z_0})}]^2},\tag{S21}$$

where  $e_s$  is the saturation vapor pressure, and  $e_a$  is the actual vapor pressure at present state.  $\rho_{\text{water}}$  is the density of water, respectively. The wind speed u (m s<sup>-1</sup>) is calculated from the housing ventilation at an assumed reference height of z that equals 2 m, with roughness height  $z_0$  is assumed to be  $2 \times 10^{-3}$  m (2 mm).

# S3.2 Simulations for poultry housing

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The third type of animal house in AMCLIM-Housing is designed specifically for poultry housing simulations. This accounts for the fact that poultry excreta are in the form of uric acid which hydrolyses to TAN much more slowly than urea (see Appendix A1). Furthermore, poultry excreta are much drier than pig excreta, so the rate of uric acid hydrolysis is also limited by the moisture levels (see Appendix A1). Housing management for poultry can also differ from other livestock. Addition of bedding materials to poultry excreta produces a solid litter. Consequently, poultry litter can be left in houses for a longer period than for other housed livestock, i.e., so called "deep litter" systems.

In AMCLIM-Housing, the water pool in poultry houses is determined by the initial water content in the excreta ( $F_{\text{excretion}}$ ), evaporation, and the cleaning function, as shown in the following equation:

$$\frac{dM_{\rm H_2O}}{dt} = \max\left(F_{\rm excretion\ water} - F_{\rm evap}, m_{\rm E}M_{\rm DM}\right) - \psi_{\rm cleaning}(t, \rm H_2O) \tag{S22}$$

where  $m_{\rm E}$  is the equilibrium moisture content of the excreta as a function of ambient temperature and humidity.  $M_{\rm DM}$  is the mass of dry matter (DM) of the excreta, which is used to determine the water at equilibrium moisture.

The moisture in poultry litter and solid manure due to evaporation cannot decline further than a threshold and will eventually reach an equilibrium state to the ambient humidity, and evaporation is assumed to stop at this point. 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 and Perry, 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. The equilibrium moisture content is calculated by the following equation (Elliott and Collins, 1982):

$$m_E = \left[ \frac{-\ln\left(1 - \frac{RH}{100}\right)}{0.0000534 \times T} \right]^{\frac{1}{1.41}}.$$
 (S23)

The high DM content of the poultry litter can result in NH<sub>4</sub><sup>+</sup> adsorption on litter solids, a process similar to NH<sub>4</sub><sup>+</sup> adsorption on soil particles (as described in Section 3.2.1.1; Equation 3.4). Due to the lack of knowledge regarding nitrogen adsorption on livestock manure, AMCLIM–Land uses a constant partitioning coefficient (*K*<sub>d</sub>) of 1.0 for all livestock (ref, Vira et al., 2020), so the amount of N adsorbed on manure solid is only dependent on the water content of the manure. Moreover, the surface of poultry excreta can dry quickly, forming a natural outer "crust" that prevents further emissions from the old litter below. The quantity of this layer is uncertain, and modelling the drying process is difficult. To simulate the NH<sub>3</sub> volatilization from poultry excreta, AMCLIM–Housing assumes an additional surface resistance of 8640 s m<sup>-1</sup> (0.1 d m<sup>-1</sup>) for litter (*R*<sub>litter</sub>). This surface resistance is derived using an inversion method as described in the previous version of AMCLIM-Poultry (Jiang et al., 2021). For deep litter system, surface resistance doubles (17280 s m<sup>-1</sup> or 0.2 d m<sup>-1</sup>) due to bedding materials added.

# S3.3 Simulations for ruminant housing

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Ruminants including cattle, sheep and goats, are typically kept in naturally ventilated animal houses as these animals have higher tolerances to cold temperatures than pigs and poultry. In AMCLIM, it is assumed that the excreta from these animals are removed from the houses on a daily basis. Meanwhile, ruminants also graze outside, which leads to the deposition of excreta on pastures. Two grazing systems are considered: year-round grazing and seasonal grazing. In the case of year-round grazing, all ruminant excreta are assumed to be deposited on pastures. For seasonal grazing, the excreta are split into two parts, with a fraction of excreta remaining in the animal houses, while the rest is left outside while grazing. The time evolution of N pools ( $M_N$ ; given in per unit area; all masses have units of g m<sup>-2</sup> if not specifically explained) in the animal houses can be modified from Equation 4.6 as follows:

$$\frac{dM_{N_i}}{dt} = (1 - f_{\text{grazing}}) F_{\text{excretN}} f_{N_i} - K_{N_i} M_{N_i} - \psi_{\text{cleaning}}(t, N_i), \tag{S24}$$

where  $f_{\text{grazing}}$  is the fraction of ruminant excreta that is deposited on pastures and is dependent on the grazing time. As described in Section 4.2,  $F_{\text{excretN}}$  is the total N excretion rate from the livestock, and  $f_{\text{N}}$  is the fraction of a N form in the excretion.  $K_{\text{N}}$  is the conversion rate (s<sup>-1</sup>) at which a N species decomposes.  $\psi_{\text{cleaning}}(t)$  represents the cleaning event of the house (see Equation 4.5).

The characteristics of ruminant excreta are similar to pigs, as they contain both urine and dung, with excreted N mainly existing as urea in urine and organic N in faeces. The differences between ruminant and pig excreta stem from the biological and behavioural features that are varied between livestock, such as urinary N concentration, faecal N content, urination and defecation volume/mass and frequency etc. Further information is given in Appendix B6. The TAN pool can be calculated by Equation 4.4. Similarly, the water pool is calculated from urination ( $F_{\text{urine}}$ ), water in faecal excreta ( $F_{\text{faecal water}}$ ), loss by evaporation of water ( $F_{\text{evap}}$ , mm s<sup>-1</sup>), and the cleaning event by the following equation:

$$\frac{dM_{\text{H}_2\text{O}}}{dt} = (1 - f_{\text{grazing}})(F_{\text{urine}} + F_{\text{faecal water}}) - F_{\text{evap}} - \psi_{\text{cleaning}}(t, H_2\text{O}). \tag{S25}$$

# S4 Nitrification process of livestock manure

Nitrification is considered to take place in soils and solid manure systems exposed to oxygen. In contrast, for liquid systems, such as slurry system or lagoon, nitrification is considered to be absent or negligible due to the high water content that reduce the oxygen availability. In the model, nitrification is included for calculating the TAN pool in solid phase manure management simulations.

A first-order reaction is used to determine nitrification (refs). The optimum nitrification rate ( $K_{\text{Knitrif,opt}}$ ) is set to be 10 % per day, and the nitrification rate  $K_{\text{nitrif}}$  is affected by temperature, water content, and pH as shown in Equation 3.6 (Parton et al., 1996, 2001). The dependence of each factor is expressed by the following equations. The temperature dependence is taken from Stange and Neue, (2009):

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$$k_{\text{nitirif,T}} = \left(\frac{t_{\text{max}} - T_{\text{gnd}}}{t_{\text{max}} - t_{\text{opt}}}\right)^{a_{\Sigma}} \exp\left(a_{\Sigma}\left(\left(\frac{t_{\text{max}} - T_{\text{gnd}}}{t_{\text{max}} - t_{\text{opt}}}\right)\right), \tag{A.17}$$

where  $T_{gnd}$  is the ground temperature. The maximum temperature ( $t_{max}$ ) and optimum temperature ( $t_{opt}$ ) for microbial activity is 313 K and 301 K, respectively.  $a_{\Sigma}$  is an empirical factor that equals to 2.4 for manure; optimum temperature is 303 K (Stange and Neue, 2009).

The water content and pH dependence are taken from the empirical function of Patron et al. (1996)

$$k_{\text{nitrif,WFPS}} = \left(\frac{w_{FPS-b}}{a-b}\right)^{d \cdot \left(\frac{b-a}{a-c}\right)} \left(\frac{w_{FPS-c}}{a-c}\right)^{d}, \tag{A.18}$$

where *WFPS* is the water-filled porosity of soil and is set to 1.0 for solid manure storage. Coefficients a, b, c and d equal to 0.60, 1.27, 0.0012 and 2.84, respectively (Parton et al., 1996).

$$k_{\text{nitrif,pH}} = 0.56 + \frac{\tan^{-1}(0.45\pi(\text{pH}-5))}{\pi}.$$
 (A.19)

Nitrification is found to taking place in soils at pH ranging between 5.5 to 10, with the optimum pH is around 8.5 (Parton et al., 1996), and the processes ceases in soils under natural pH less than 5.0 (Parton et al., 1996). In AMCLIM-Land, the pH dependence for nitrification rate is a trigonometric function from Patron et al. (1996).

#### S5 Concentrations of nitrogen species at surface

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Volatilization take place at the emitting surface, which is primarily driven by the concentrations at the surface. For simulating NH<sub>3</sub> emissions from solid manure storage, the processes are similar to the land simulations (Jiang et al., submitted 2024). TAN is assumed to be evenly distributed in the stored manure, so the TAN concentration represents the concentration of the bulk manure ([TAN(aq)]<sub>bulk</sub>). TAN is transferred from the manure to a source layer at the surface through diffusions. The diffusion is in aqueous phase considering the water content and is constrained by manure resistance. Manure resistance is determined by dividing the thickness of the surface layer which has a maximum thickness of 2 cm by the aqueous diffusivity of NH<sub>4</sub><sup>+</sup>. The upward diffusive fluxes are equal to the volatilization flux (as there is no runoff for housing and storage). Therefore, the TAN concentration at the manure surface can be solved by the following equation:

$$[TAN(aq)]_{srf} = [TAN(aq)]_{bulk} \cdot \frac{\left(\frac{1}{R_{manure,aq}}\right) + \frac{\chi_{in}}{R_{store}}}{\frac{1}{R_{manure,aq}} + \frac{K_{NH_3}}{R_{store}}}.$$
(A.44)

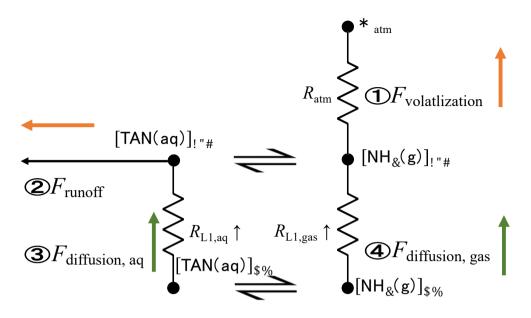


Figure S2. Sketch of the physical transport for nitrogen species (TAN as an example) in the top soil layer in AMCLIM-Land. Upward diffusions including aqueous and gaseous diffusive flux are equivalent to the surface runoff and volatilization to satisfy mass conservation (process 1+2=3+4; the sum of the fluxes represented by orange arrows = the sum of the fluxes represented green arrows).

# S6 Model setup for global simulations for livestock farming

#### S6.1 Housing environments and housing density

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There are two housing systems considered in AMCLIM-Housing: fully enclosed houses (with forced heating and ventilation) and partially enclosed houses as described in Section XXX. The inside conditions of animal houses significantly influence the NH<sub>3</sub> emission from livestock housing as they can be very different from the natural environment, with indoor temperature being the most prominent environmental factor. Pigs and poultry have a lower critical temperature (i.e., the minimum managed temperature for optimum chicken performance) of approximately 16–20 °C (Gyldenkærne, 2005). Therefore, pigs and poultry from commercial production systems that are intensively managed (e.g., industrial pigs, broilers and layers) are typically kept in insulated buildings equipped with forced heating and ventilation systems. These systems help maintain the ambient temperature within a recommended range throughout the year as far as feasible (Seedorf et al., 1998). Heating is used on cold days when the temperature is low, while ventilation is used to cool down the house when the temperature is high. Fully enclosed houses require a minimum level of ventilation to remove odours and emissions like NH<sub>3</sub> from the house, which aims to maintain a healthy environment for the animal growth. However, the ventilation should also be below a certain rate to avoid causing an induced draft in the house. For intermediate and backyard production systems,

pigs and poultry are kept in barns that are naturally ventilated. These barns have indoor environments that are closer to the natural environments, with slightly higher temperatures than outdoor temperatures due to the warmth generated by the animals, and local materials are used to block wind and to warm the buildings in cold days.

In AMCLIM-Housing, the indoor temperature and ventilation of animal houses are modelled using a set of empirically derived relationships in relation to the outdoor temperature. These relationships are based on data from the Animal Feeding Operations (AFOs) dataset by the US Environmental Protection Agency (EPA, 2012) and theoretical parameterizations of indoor environments by Gyldenkærne et al. (2005). These relationships can vary between livestock sectors and production systems as each production system of livestock has a corresponding housing system and house type in the global simulations. Table XXX lists the housing system and house type of livestock by production systems used in AMCLIM-Housing.

# 230 Table S1. Housing systems and house types for livestock in AMCLIM-Housing.

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Production system	Housing system	House type
Pigs		
Industrial	Fully enclosed house	Houses with slatted floors and storage pits
Intermediate	Naturally ventilated house	Normal barns
Backyard	Naturally ventilated house	Normal barns
Poultry		
Broiler	Fully enclosed house	Poultry houses
Layer	Fully enclosed house	Poultry houses
Backyard	Naturally ventilated house	Poultry houses
Ruminants		
Mixed	Naturally ventilated house	Normal barns

For the enclosed houses with heating and ventilation systems for pigs, the parameterizations of housing environments are taken from Gyldenkærne (2005), as shown in Figure A3. The indoor temperature ( $T_{in}$ ,  ${}^{o}$ C) is a function of outside temperature ( $T_{out}$ ,  ${}^{o}$ C), as the following:

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$$T_{\text{in}} = \begin{cases} T_{\text{rec}} + \Delta T_{\text{low}} \times (T_{\text{out}} - T_{\text{min}}), & \text{if } T_{\text{out}} \leq T_{\text{min}} \\ T_{\text{rec}}, & \text{if } T_{\text{min}} < T_{\text{out}} \leq T_{\text{max}} \\ T_{\text{rec}} + \Delta T_{\text{high}} \times (T_{\text{out}} - T_{\text{max}}), & \text{if } T_{\text{max}} < T_{\text{out}} \end{cases}$$
(A.62)

where  $T_{\text{rec}}$  is the recommended temperature (20 °C),  $\Delta T_{\text{low}}$  is the temperature dependency (0.5 °C °C<sup>-1</sup>) for temperatures below  $T_{\text{min}}$  (0 °C),  $\Delta T_{\text{high}}$  is the temperature dependence (1.0 °C °C<sup>-1</sup>) above  $T_{\text{max}}$  (12.5 °C).

For the enclosed poultry houses, the temperature relationships are derived from the USEPA AFO dataset as the follows (as shown in Fig A3):

$$T_{\rm in} = \begin{cases} 2.0 \times 10^{-4} T_{\rm out}^{3} + 1.0 \times 10^{-3} T_{\rm out}^{2} + 2.4 \times 10^{-2} T_{\rm out} + 22.1, \text{ for broilers} \\ 1.4 \times 10^{-4} T_{\rm out}^{3} + 2.3 \times 10^{-3} T_{\rm out}^{2} + 1.1 \times 10^{-2} T_{\rm out} + 23.8, \text{ for layers} \end{cases}$$
(A.63)

The ventilation (V<sub>in</sub>, m s<sup>-1</sup>) of the enclosed animal houses calculated as follows (as shown in Fig A3):

$$V_{\rm in} = \begin{cases} V_{\rm min}, & \text{if } T_{\rm out} \leq T_{\rm min} \\ V_{\rm min} + T_{\rm out} \times (\frac{(V_{\rm max} - V_{\rm min})}{(T_{\rm max} - T_{\rm min})}), & \text{if } T_{\rm min} < T_{\rm out} \leq T_{\rm max}, \\ V_{\rm max}, & \text{if } T_{\rm max} < T_{\rm out} \end{cases}$$
(A.64)

where  $V_{\rm min}$  is the minimum ventilation (0.2 m s<sup>-1</sup>), and  $V_{\rm max}$  is the maximum ventilation rate (0.38 m s<sup>-1</sup> for pigs; 0.40 m s<sup>-1</sup> for poultry). It is worth noting that the unit of ventilation is expressed in meter per second, which should be distinguished from the ventilation rate used in Equation 4.2 for conceptualising the indoor NH<sub>3</sub> concentration of animal houses.

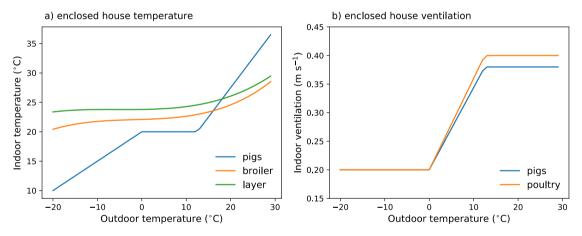


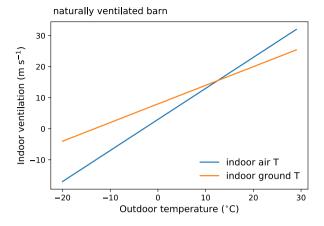
Figure S3. Modelled indoor temperature and ventilation of fully enclosed animal houses for pigs and poultry in relation to outdoor temperature.

For naturally ventilated barns where ruminants, intermediate pigs, and backyard pigs and poultry, the relationship between indoor temperature and the outdoor temperature is expressed as follows as shown in Figure A4:

$$T_{\rm in} = T_{\rm out} + D_{\rm temp},\tag{A.65}$$

$$T_{\text{floor}} = T_{\text{out}} + 0.4 \times (T_{\text{rec}} - T_{\text{out}}), \tag{A.66}$$

where  $D_{\text{temp}}$  is the temperature difference between indoor and outdoor temperature due to the warmth generated by animals (3 °C) and  $T_{floor}$  is floor temperature.



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Figure S4. Modelled indoor air and ground temperature of naturally ventilated animal barns in relation to outdoor temperature.

The ventilation in the barns is related to the wind speed outside ( $u_{out}$ , m s<sup>-1</sup>), which is expressed by the following equation:

$$V_{\rm in} = (1 - f_{\rm blocking})u_{\rm out},\tag{A.67}$$

where  $f_{\text{blocking}}$  is a blocking factor due to mechanical blocking, which is larger in cold days and smaller in warm days.

$$f_{\text{blocking}} = \begin{cases} 0.2, & \text{if } T_{\text{out}} > T_{\text{floor}} - D_{\text{temp}} \\ 0.8, & \text{if } T_{\text{out}} \le T_{\text{floor}} - D_{\text{temp}} \end{cases}$$
(A.68)

Housing density varies depending on the livestock and production system. Industrial pigs are assumed to be housed at a typical density of 120 kg liveweight per square meter (Lim et al., 2010). By comparison, intermediate and backyard pigs are housed at lower densities than the industrial production system, with assumed values of 80 and 60 kg liveweight per square meter, respectively. Regarding poultry housing, the assumed density for broilers and layers are 15 and 30 birds per square metre, respectively (Cortus et al., 2010a, b; Wang et al., 2010). Backyard poultry are less densely housed than broilers and layers, with an assumed density of four birds per square meter. For cattle, the housing density of 100 kg liveweight per square meter is assumed for beef, 80 kg liveweight per square meter for all dairy, and 150 kg liveweight per square meter for feedlot cattle. The housing area is calculated accordingly by the following equation:

$$S_{\text{house}} = \begin{cases} \frac{n_i m_i}{de n_{\text{housing}}}, & \text{if i is pig} \\ \frac{n_i}{de n_{\text{housing}}}, & \text{if i is poultry'} \end{cases}$$
(4.21)

where  $n_i$  and  $m_i$  are the number of animals and average body weight (kg head-1), and  $den_{\text{housing}}$  is the housing density of the livestock (kg animal per m<sup>2</sup> for pigs and number of animal per m<sup>2</sup> for chicken). It is worth noting that pig houses with slatted floor and pit have two NH<sub>3</sub>-emitting surfaces, so the slats areas ( $S_{\text{slats}}$ , m<sup>2</sup>) and pit areas ( $S_{\text{pit}}$ , m<sup>2</sup>) are calculated separately:

$$\begin{cases}
S_{\text{slats}} = (1 - f_{\text{gap}}) S_{\text{housing}} \\
S_{\text{pit}} = f_{\text{pit}} S_{\text{housing}}
\end{cases},$$
(4.22)

where  $f_{gap}$  is the fraction of gap space in the slats (assumed to be 0.2, i.e., 20 % of gap space, for global simulation), and  $f_{pit}$  is the relative area of the pit to the housing area (set to be 1.0 in AMCLIM-Housing, meaning that the pit surface has an equivalent size as the area of the house).

To estimate housing NH<sub>3</sub> emissions in global simulations, it is assumed that indoor and atmospheric NH<sub>3</sub> concentrations are negligible, given that animal houses are significant NH<sub>3</sub> sources and their surface concentrations are much higher than indoor and outdoor concentrations. However, as the global volume of animal houses is uncertain (as described in Equation 4.2), the calculation of NH<sub>3</sub> emissions is simplified by using the following equation:

$$F_{\rm NH_3} = \frac{\chi_{\rm srf}}{R_{\rm G,house}}.$$
 (4.23)

# S6.2 Manure storage and manure application

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Manure storage, land application of manure, and housing are closely interrelated. In particular, there are several management systems related to housing that should be specifically pointed out. In houses with slatted floor and pits, manure can be stored in the house pit either for long-term or short-term periods. For long-term pit storage, excreta are assumed in AMCLIM to be stored for two months (60 days) before being applied to the land. For short-term storage, excreta are removed from the pit daily and stored in a separate storage unit (also for the naturally ventilated barns) before ultimately being applied to the land. The specific in-situ storage management systems are determined by the MMS information in the GLEAM database.

For broiler housing with litter management, AMCLIM assumes that excretions remain in the houses for the entire year, being applied to land once being removed. It should be noted that the NH<sub>3</sub> emissions from in-situ storage are counted as part of housing emissions. In contrast, naturally ventilated barns are assumed to be cleaned daily so that excreta are removed from the house and are stored separately.

Livestock excreta removed from the houses are typically stored for a certain period before being applied to the land.

However, the area of the storage facilities is uncertain. In the AMCLIM model, it is assumed that the area for manure storage (S<sub>storage</sub>) is proportional to the housing area, which is expressed as:

$$S_{\text{storage}} = f_{\text{MMS}} f_{\text{store-housing}} S_{\text{housing}}, \tag{4.24}$$

where  $f_{\text{MMS}}$  is the fraction of manure that is removed for separate storage as part of the MMS. The ratio of storage area to housing area ( $f_{\text{store-housing}}$ ) varies depending on the specific management system. The ratio is set to be 0.5 for liquid manure storage and 0.25 for solid manure storage, and 2.5 for lagoon management, given that liquid manure storage requires a larger area because the volumes are larger than those of solid manure.

In AMCLIM, it is assumed that the stored manure is kept for 180 days and then is applied to the land twice a year during the spring and autumn planting seasons, respectively. The application date is based on the average value of the crop calendars for 18 spring crops and 4 winter crops (see Section 2.4.1, 3.2.3.2 and Appendix B8). For slurry application, the application rate is assumed to be 3 mm of slurry, which is equivalent to a recommended rate of 30 tons per hectare. For solid manure

application, a moderate fertilization rate of 10 tons per hectare is used. The N pools and the water pool are calculated accordingly. It should be noted that all stored manure, with the exception of manure in lagoons, is assumed to be applied to agricultural lands. The lagoon system is a small fraction among all management systems, and manure in this system is assumed not to be applied to land but to be kept in the lagoons in AMCLIM.

# 310 S6.3 Grazing density

The grazing density for all cattle is set at 2500 square meter per head (equivalent to four animals per hectare; Saarijärvi et al., 2006; Saarijärvi and Virkajärvi, 2009). For sheep and goats, a housing density of 50 kg liveweight per square meter is assumed and a grazing density of 400 square meter per head (equivalent to 25 animals per hectare). The housing areas are calculated by Equation 4.21 as for pigs. For grazing, the area ( $S_{\text{grazing}}$ ,  $m^2$ ) can be calculated by the following equation:

$$S_{\text{grazing}} = n_{\text{i}} de n_{\text{grazing}}, \tag{5.6}$$

note that grazing "density" ( $den_{grazing}$ , m<sup>2</sup> head<sup>-1</sup>) has a different unit from housing density ( $den_{housing}$ , kg animal weight m<sup>-2</sup>) as mentioned. It is important to clarify that the source areas of NH<sub>3</sub> emissions from grazing are not equivalent to the grazing area. Saarijärvi et al. (2006) have shown that the annual average surface coverage of urine and dung on a grazing field is 17 % and 4 %, respectively. In AMCLIM, the source areas of NH<sub>3</sub> emissions from urine patch ( $S_{urine\ patch}$ ) and dung pat ( $S_{dung\ pat}$ ) can be expressed as follows:

can be expressed as follows.

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$$S_{\text{urine patch}} = f_{\text{urine}} S_{\text{grazing}},$$
 (5.7)

$$S_{\text{dung}} = f_{\text{dung}} S_{\text{grazing}}, \tag{5.8}$$

where  $f_{\text{urine}}$  is 0.17 and  $f_{\text{dung}}$  is 0.04. The areas for dung-only and dung mixtures in the dung pat scheme are the same, which accounts for 2 % of the total grazing area.

#### 325 S7 Site simulations of layer housing using AMCLIM

Figures 4.3 shows the simulated NH<sub>3</sub> emissions and indoor concentrations of a layer house compared with the measurements, along with indoor conditions and modelled N species (other simulations shown in Appendix D1). The indoor environments of the layer house are similar to the pig house, with temperature being largely maintained between 20 to 30 °C throughout the year and ventilation working intensively in hot summer. Relative humidity inside the layer house shows strong daily variations, ranging between 40 to 80 %.

The simulated period is from 15 March 2008 to 15 March 2009. The house was fully occupied by more than 90 000 layers for most of the time and was only emptied once (on 04 April 2008) for three weeks. Overall, the model captures the major changes of NH<sub>3</sub> emissions and indoor concentrations well over the simulation period. High emissions occur in summer as the ventilation increases, with the emissions peaking in early June 2008. The maximum daily NH<sub>3</sub> emission is more than 150 kg d<sup>-1</sup>, and AMCLIM–Housing roughly reproduces this value but with a lag of ~5 days, in the timing of the peak in early June 2008. The average daily emission of NH<sub>3</sub> estimated by the model is 63.6 kg d<sup>-1</sup> (when measurements available; 62.4 kg d<sup>-1</sup>

for the entire simulation), compared to 54.4 kg d<sup>-1</sup> reported by the measurements. Approximately 34 % of total excreted N is lost due to NH<sub>3</sub> emissions according to the simulation.

The indoor NH<sub>3</sub> concentrations show an opposite trend to the emission, which is inversely related to the ventilation. The indoor NH<sub>3</sub> level is typically lower than 10 mg m<sup>-3</sup> when the airflow rate is high in summer and was much higher in the winter when the ventilation decreases, reaching to around 30 mg m<sup>-3</sup> from November 2008 to February 2009. AMCLIM–Housing replicates the measured indoor NH<sub>3</sub> concentration well. However, the model largely underestimates both the emissions and concentrations in the first simulated month before the house is emptied. The NH<sub>3</sub> concentration at the surface is much higher than the indoor concentrations, ranging from 0.5 to 4.0 g m<sup>-3</sup> (500 to 4000 mg m<sup>-3</sup>), which generates a concentration gradient that drives the emission fluxes.

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As shown Figure 4.3c, the uric acid pool in the excreta gradually increases in the first three months of the simulation and then generally stabilizes in the remaining period. There are two decreases in the simulated uric acid pool, with the first drop due to the emptying of the house in early April 2008 and the second due to a sharp increase of indoor temperature that accelerates the hydrolysis process in late May 2008. By comparison, the TAN pool accumulates throughout the year, building up to about 7 g N m<sup>-2</sup> at the end of the simulation. It is notable that the variations in surface concentration of NH<sub>3</sub> are similar to those in NH<sub>3</sub> emissions. This is because the litter resistance (8640 s m<sup>-1</sup>) is much larger than the housing resistance that range between 200 to 600 s m<sup>-1</sup>. As a result, the total resistances show small variability. The NH<sub>3</sub> emissions are mainly constrained by the litter resistance, so the emissions and concentrations broadly display the same feature.

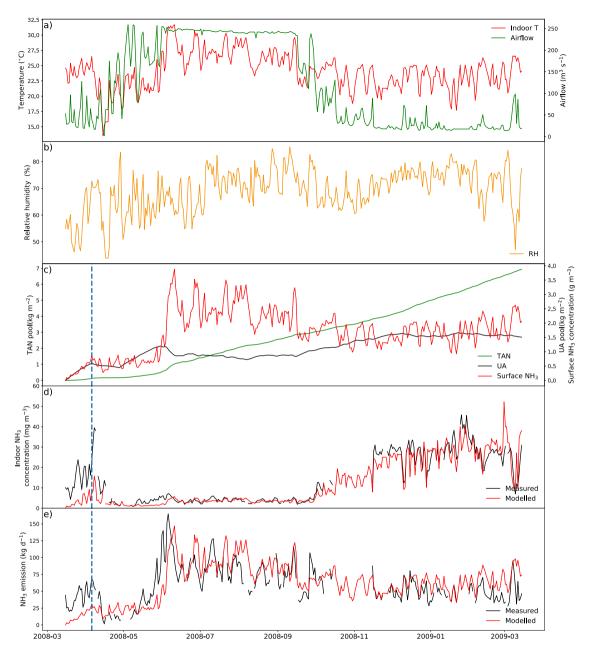


Figure S5. Site simulations of House A in a layer farm at site NC2B, Nash, North Carolina, from 15 March 2008 to 15 March 2009.

(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. (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. Vertical blue dashed lines refer to emptying of the house.

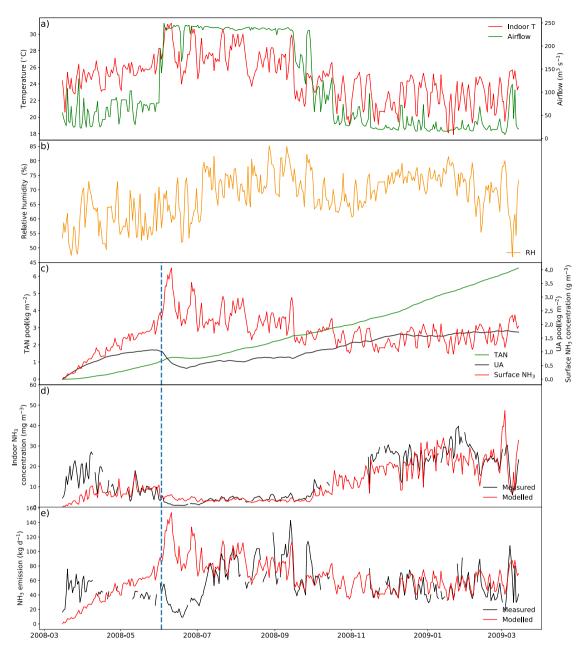


Figure S6. Same as Figure S5 but for House B at site NC2B.

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