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Estimate of changes in agricultural terrestrial nitrogen pathways and ammonia emissions from 1850 to present in the Community Earth System Model

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Table S1 Site description and measurement method used to compare with model prediction from manure

Site Location	Source	Site Description	#	Date of measurement	Average Temperature (°C)
Texas, USA	Todd et al., 2007	Commercial beef cattle feedyard.	1	August 2002	25
			2	July 2003	28
			3	June 2004	23
			4	January 2003	1
			5	February 2004	2
The Netherlands	Bussink, 1992	Dairy cow grazing. Ryegrass.	6	May 1987	13
			7	June 1987	15
			8	July 1987	17
			9	August 1987	18
			10	September 1987	14
			11	October 1987	11
			12	May 1988	12
			13	June 1988	14
			14	July 1988	16
			15	August 1988	17
			16	September 1988	14
			17	October 1988	11
			18	September 1989	12
The Netherlands	Bussink, 1994	Dairy cow grazing. Ryegrass.	19	May 1989	10
			20	June 1989	11
			21	July 1989	13
			22	August 1989	15
UK	Jarvis et al., 1989	Beef cattle grazing land. Ryegrass	23	May 1987	13
			24	June 1987	16
			25	July 1987	22
			26	August 1987	21
			27	September 1987	19
			28	October 1987	15
			29	November 1987	13
Texas, USA	Flesch et al., 2007	Commercial beef cattle feedyard.	30	June 2004	23
			31	April 2005	20
Alabama, USA	Mulvaney et al., 2008	Dairy cattle pasture land.	32	June 2003	25
			33	September 2003	22
			34	January 2004	10
			35	April 2004	20
Switzerland	Spirig et al., 2010	Cattle slurry, managed grassland	36	July 2006	19
			37	September 2006	15
			38	October 2006	9
			39	April 2007	9
			40	July 2007	19
			41	October 2007	9

Spain	Sanz et al., 2010	Pig slurry, arable land	42	September 2006	20
Switzerland	Sintermann et al., 2011	Cattle slurry, arable/grass	43 44	August 2009 August 2009	18 18

Table S2 Site description and measurement method used to compare with model prediction from fertilizer

Site Location	Source	Site Description	#	Year	Lat.
Canterbury, NZ	Black et al., 1985	Pasture	1	1983	-43
Canterbury, NZ	Black et al., 1985	Pasture	2	1984	-43
Canterbury, NZ	Black et al., 1989	Sown wheat	3	1985	-43
Queensland, Australia	Catchpoole et al., 1983	Pasture	4	1982	-28
Texas, USA	Hargrove & Kissel, 1979	Turf	5	1976	33
Illinois, USA	Goebes et al., 2003	Cropland	6	2000	38
Georgia, USA	Vaio et al., 2008	Pasture	7	2004	31
Kentucky, USA	Bowman et al., 1987	Turf	8	1985	37
Ontario, Canada	Sheard & Beauchamp, 1985	Turf	9	1985	43
Vancouver, Canada	Nason et al., 1998	Forest	10	1986	50

Bounds for the sensitivity experiments:

Here we discuss the bounds chosen for the various sensitivity experiments (Table 1 and Table 2). In the first set of experiments on the manure (EX1m, EX2m) we control for the mechanical mixing of manure into the soil. Estimates for the diffusivity of bioturbation alone vary by an order of magnitude (Koven et al., 2013) depending on location. Tillage, grazing animals and other agriculture practices would also be expected to impact this mixing rate. Therefore we set large bounds on the timescale for this coefficient coefficient at 100 and 750 days, with the default value set at 365 days in the control experiment. Experiments EX3 and EX4 modified the adjustment time of the water within the TAN pool to the soil water for both the fertilizer and manure pools. Very little available guidance is available for choosing this parameter and therefore we varied it from diurnal (1 day) to synoptic timescales (10 days). Reasonable bounds for variations in pH are approximately between 6 and 8 (EX5, EX6, EX7). In Eghball et al. (2000) the majority of the reported measurements of pH for beef cattle feedlot manure are between 7 and 8, although in one case a pH of 8.8 was measured. The recommended pH for various crops ranges from approximately 5.8 to 7.0 depending on the crop (e.g., http://onondaga.cce.cornell.edu/resources/soil-ph-for-field-crops). In EX7 we use the ISRIC-WISE dataset (Batjes, 2005) imported dataset for soil pH, but this dataset does not account for pH changes in agriculturally amended soils. The canopy capture fraction (EX8, EX9) can vary considerably depending on vegetation type and season. Here we bounded the sensitivity calculations at canopy capture fractions of 0.4 and 0.8. Note, however, ammonia emissions are linearly dependent on this parameter (equation 8) so additional sensitivity tests are not warranted. The background NH_3 concentration has considerable spatial and temporal heterogeneity (EX10, EX11). To investigate the effects of this, uncertainty bounds were set for background ammonia concentrations at 0.1 and 1 μg m^{-3} for the lower and upper bound as representative of clean and moderately polluted sites (Warner et al., 2016). The sensitivity to the H2O depth is tested in simulations EX12 and EX13. The H2O depth is essentially taken as the mixing depth of the manure or fertilizer, the depth to which the nitrogen equilibrates with the soil water. The bounds are picked as correspond to shallow mixing (2 cm) and deeper mixing (10 cm). The diffusion timescale for the mixing of ammonia into the soils (K_D) is varied in simulations EX14 and EX15. This timescale depends on the water content, the base diffusion rate, the soil porosity, and a length scale of which the diffusion operates over (Table S1, supplement). While the water content is determined internally within the CLM all the other factors are set globally. We take ± 50% as a reasonable bound on K_D. The maximum rate of nitrification (r_{max}) is taken from Parton et al, 2001. In sensitivity experiments EX16 and EX17 we take ± 50% as reasonable bounds on this parameter. Sensitivities for experiments EX18m, EX18f, EX19f and EX20f varying the characteristics of manure or fertilizer input are given in the text.