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Simultaneous measurements of ammonia volatilisation and deposition at a beef feedlot

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Abstract. The nitrogen (N) excreted at intensive livestock operations is vulnerable to volatilisation, and, subsequently, may form a source of indirect nitrous oxide (N_2O) emissions. The present study simultaneously investigated volatilisation and deposition of N at a beef feedlot, semi-continuously over a 129-day period. These data were examined relative to pen manure parameters, management statistics and emission-inventory calculation protocols. Volatilisation measurements were conducted using a single, heated air-sampling inlet, centrally located in a feedlot pen area, with real time concentration analysis via cavity ring-down spectroscopy and backward Lagrangian stochastic (bLS) modelling. Net deposited mineral-N was determined via two transects of soil-deposition traps, with samples collected and re-deployed every 2 weeks. Total volatilised ammonia amounted to 210 tonnes of NH_3-N (127 g/animal.day), suggesting that the inventory volatilisation factor probably underestimated volatilisation in this case (inventory, 30% of excreted N; 65 g N volatilised/animal.day; a value of ~60% of excreted N is indicated). Temperature contrast between the manure and air was observed to play a significant role in the rate of emission ($R^2 = 0.38$; 0.46 Kendall's tau; $P < 0.05$). Net deposition within 600 m of the pen boundary represented only 1.7% to 3% of volatilised NH_4^+-N, between 3.6 and 6.7 tonnes N. Beyond this distance, deposition approached background rates (~0.4 kg N/ha.year).

Additional keywords: manure, micrometeorology.

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Introduction

A wide range of human activities result in ammonia volatilisation and are therefore regarded as indirect sources of greenhouse-gas (GHG) emissions (Intergovernmental Panel on Climate Change 2006). These activities include animal production (intensive and extensive), sewage treatment, and manure or inorganic-fertiliser application to land. About 65% of atmospheric ammonia (NH_3) originates from livestock manure (National Research Council 2002). When beef cattle are involved, much of this NH_3 is derived via the hydrolysis of the urea in urine, the N form that makes up ~50% of the N excreted (Mosier *et al.* 1973).

Estimates of losses of excreted N from feedlot cattle via NH_3 volatilisation vary widely. An early estimate suggested losses of ~50% due to runoff, volatilisation and denitrification before manure removal from the pen (Eghball and Power 1994). More recent measurement of NH_3 volatilisation from an Australian feedlot calculated that losses amounted to ~60% of N excreted (Denmead *et al.* 2008), and greater than 90% in another study at the same location (Loh *et al.* 2008; both >60% of dietary N). Measurements of NH_3 volatilisation from a United States feedlot (Texas) indicated losses of ~65% (Flesch *et al.* 2007), and 19–85% of dietary N (Todd *et al.* 2011).

Limited flux data from intensive livestock-production systems is available. An early study of NH_3 volatilisation from beef feedlots focussed on air concentrations of NH_3, from immediately adjacent to beef feedlots to up to a kilometre distant (McGinn *et al.* 2003). One study of a Texan beef feedlot found annual NH_3 volatilisation of ~19.3 kg NH_3 per head (39 days of measurement spread throughout 3 years; Todd *et al.* 2008*a*). Volatilisation from a southern Alberta feedlot was observed to be ~89 kg NH_3 per head per year (Staebler *et al.* 2009) or 53 kg per head per year (McGinn *et al.* 2007). A study involving steers undergoing backgrounding and finishing on varied diets at a Canadian feedlot observed NH_3 volatilisation of 3.5–62.8 g NH_3 per head per day (annualised to 1.6–28 kg NH_3 per head per year; Koenig *et al.* 2013). A recent 8-day campaign in Victoria (Australia) observed higher rates of volatilisation, equivalent to ~43 kg NH_3 per head per year (Bai *et al.* 2015).

Ultimately, much of the volatilised NH_3 is deposited from the atmosphere via two processes, namely wet and dry deposition. Wet deposition occurs via in-cloud processes (rain clouds) or through wash out of the atmosphere via rain, and subsequent deposition to the land surface and the surfaces of plants. Dry deposition occurs where there are low land-surface NH_3 concentrations relative to those of the air (forming a gradient), the process representing the opposite process to volatilisation (Asman 1998). The relative importance of these processes appears clear (Krupa 2003): wet deposition dominates where atmospheric

concentrations are low, and dry deposition dominates where these concentrations are high, e.g. close to a major source of contamination.

Several published studies have suggested that some NH_3 deposition occurs close to the source, whereas much of the volatilised NH_3 is advected away. As little as 10% of volatilised NH_3 was deposited (dry deposition) within 4 km of the source in one study (Staebler *et al.* 2009). A preceding study found that only 3–10% of volatilised NH_3 from a poultry shed was deposited within 300 m of the source (Fowler *et al.* 1998). In these scenarios, the advected, possibly dilute, plume of dispersing NH_3 may be re-deposited to the wider landscape. Similarly, a recent publication suggests that ~8% of estimated volatilisation was deposited within 1 km of an Australian beef feedlot (Shen *et al.* 2016). Shen *et al.* (2016) based the magnitude of source NH_3 volatilisation on a previous 8-day measurement of pen NH_3 volatilisation and a 10-day measurement of stockpile NH_3-volatilisation measurements from a beef feedlot (Bai *et al.* 2015). To our knowledge, simultaneous measurement of volatilisation and deposition has been conducted only once previously from a beef feedlot (McGinn *et al.* 2016). These authors found that ~14% of emitted NH_3 was deposited within 500 m of an Alberta (Canada) feedlot. Another study used on-going NH_3-volatilisation measurements via open-path laser and three flights of an airborne analyser through the plume to calculate estimates of both volatilisation fluxes and dry deposition (Staebler *et al.* 2009), finding that ~90% of feedlot-emitted NH_3 was advected away.

However, close to the source, the rates of deposition can be high. Within 0–700 m of various volatilisation sources, deposition rates of up to 254 kg N per ha have been observed (Berendse *et al.* 1988; McGinn *et al.* 2003; Todd *et al.* 2008*a*; Staebler *et al.* 2009).

The relationship between deposition to the soil and volatilisation-flux estimates is unclear, as simultaneous volatilisation and deposition measurements are rare in the literature. Our study differed from those summarised here, in that deposition to the landscape surrounding the feedlot and volatilisation from a human activity (intensive beef cattle management at a feedlot) were simultaneously measured for a moderately long period (continuous for 129 days, more than 50% longer than the measurement period employed by McGinn *et al.* 2016). Additionally, these deposition measurements are collected in a southern hemisphere environment (Queensland, Australia). In this regard, our study appears to be unique. Our hypothesis was that only a small proportion of volatilised NH_3 from a feedlot source is deposited in close proximity to its boundaries, and that volatilisation losses are in agreement with recent measurements (50–90+ % of excreted N) rather than the inventory estimate (30% of excreted N; Intergovernmental Panel on Climate Change 2006).

Materials and methods

Site selection

Measurements were conducted at an Australian feedlot, located on the Darling Downs in Queensland. This location has an average summer-dominated rainfall of 634 mm, and an annual average temperature of 25°C. Cattle on-feed were recorded daily for the study period (a short trial period from 1 February 2013 to 2 March 2013 and a longer campaign from 12 February 2014 to 17 June 2014) by the operators and the data were made available for the purposes of the present study (average 12 779, minimum 10 201, maximum 15 373 cattle; average cattle mass 430 kg). Feed was delivered in the morning and was available throughout the day. Livestock were fed a total mixed ration, consisting primarily of sorghum, forage and white fluffy cotton seed, together with additives. The formulated crude protein of the ration averaged 9.6%.

The enterprise is sited on a uniform self-mulching expanding clay soil, typical of the highly productive broadacre cropping areas of the Darling Downs (Vertisol soil, Soil Survey Staff 1998; or Black Vertosol, Isbell 2002).

Site instrumentation and calculated manure condition

Instrumentation was largely located in a clear area among the pens themselves (Fig. 1), and within 60 m of the calculated centroid of the pen area. Wind data were monitored using a sonic anemometer (CSAT 3d, Campbell Scientific; https://www.campbellsci.com.au/csat3, accessed October 2017; 1.5 m height), air temperature logged from a shielded probe (HMP45 C, Vaisala; www.vaisala.com, accessed October 2017), acoustic air temperature was provided via the sonic anemometer (Flesch *et al.* 2007), and rainfall was recorded via a tipping bucket rain gauge (odysseydatarecording.com/, accessed October 2017). Additional data for the site were reported in another study conducted at the site over an overlapping period where manure emissions were measured and modelled (Redding *et al.* 2015). The present study also refers to publicly available

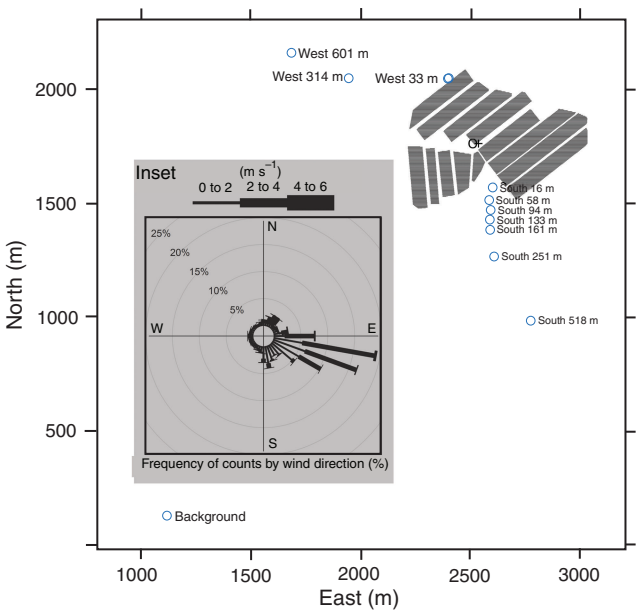

Fig. 1. Diagram of feedlot-site layout, the location of deposition traps, and the sampler intake ('O'). The intake, sonic anemometer, temperature, and rainfall monitoring equipment were located close to the anemometer, within 60 m of the centroid of the pen area ('+'). The windrose for the period is included as an inset, while the shaded portion is the pen area.

air temperature, rainfall, humidity and evapo-transpiration data for the site (www.bom.gov.au, accessed October 2017).

Ammonia sampling and measurement

A filtered-air sampling intake (Mykrolis cartridge filter, catalogue number WGFG21KP3; www.entegris.com, accessed October 2017) was located at the height of the top of the pen rail (1.5 m) within 60 m of the centroid of the total pen area (shaded portions, Fig. 1). A stainless-steel sample line led 10 m to the instrument enclosure. Nickel–chromium wire was coiled around this stainless-steel intake line (with an outer layer of domestic pipe lagging), and the air stream temperature was maintained at 60°C via controlled 24 V current (novusautomation.co.uk; accessed October 2017), so as to decrease sorption of NH_3 to the walls of the sampling tube.

Sample air was drawn into the intake at a flow rate of 2.5 L/min (flow rates controlled by Alicat MC series 10-L capacity; www.alicat.com; gas volumes standardised to 101.325 kPa and 25°C), via a diaphragm vacuum pump (12 V KNF; www.knf.com). This flow rate produced a sample residence time in the 4-mm (internal diameter) tubing of ~3 s.

Analyses for NH_3 were conducted using a Cavity Ring Down Spectrophotometer (Picarro Model 2130; www.picarro.com, accessed October 2017; rolling 30-s averages).

The backward Lagrangian stochastic (bLs) technique was applied to determine the flux of NH_3 (Flesch and Wilson 2005), using the data from the intake and the sonic anemometer, in conjunction with the Windtrax model (Crenna *et al.* 2008). Micrometeorological flux calculations were conducted using the half-hourly gas analyses and wind statistics.

Standard NH_3 gas releases of four concentrations (< 0.005, 1.30, 14.0, 19.0 ml NH_3 gas per m^3 of air, equivalent to <5, 1300, 14 000, and 19 000 ppb respectively; concentrations determined by instrument-grade air and permeation tubes available) were used to determine what lag correction was required to account for tube transit time.

Standard rejection criteria (Flesch *et al.* 2005, 2007; Loh *et al.* 2008) were applied, where the friction velocity (u^*) fell below 0.15 m/s, the Obukhov length (L) was between +10 and –10 m, and where the estimated roughness height exceeded the sampler height. Data with inappropriate wind directions for the intake layout were removed from the dataset, which generally removed Windtrax-calculated emission estimates with high standard deviations.

Deposition traps

Net deposition (deposition less re-volatilisation) to the soil was investigated via soil traps, using a method similar to that described previously (McGinn *et al.* 2003), although without a rain shelter. These low-profile traps were designed to have a minimal influence on net deposition conditions relative to their surrounds and contained the same soil type as the surrounding area.

Polypropylene lids with an internal 0.0478-m radius were used as a soil reservoir (affixed with glue to a ceramic tile to provide stability in the field).

A mass of 15 kg of soil was collected from adjacent to the western 601-m transect site (Fig. 1), from the surface to a depth of 10 mm. This soil was sieved to pass a 2-mm-diameter aperture, but was retained in a field-moist state at room temperature in a well aerated container.

During the continuous NH_3 volatilisation monitoring period (~5 months), seven trap deployments (each of ~3 weeks) were conducted. Where rainfall occurred, the deployment period was curtailed and the traps were collected within 18 h. A mass of 62 g (oven-dry equivalent, but in the moist condition described) was deployed in each deposition trap, into the field. Two traps were placed at each of the western and southern transect sites, an additional three at the background site, and a further sample was placed in a jar in the laboratory and maintained at 25°C for the duration of the deployment (Fig. 1). A sample of the soil deployed to the field was also retained in a sealed vial and analysed at the same time as the samples recovered from the field.

The location of deposition traps at the site (Fig. 1) was restricted by the normal operation of this feedlot enterprise. Sites were selected to allow representation of both the dominant wind direction, and a wind direction representative of a less common orientation. Trap sites were maintained in a bare fallow state.

At the end of each deployment, the soil samples were recovered from the deposition trap, and immediately bottled in the field. The samples were stored frozen in the laboratory until analysis via 2 M KCl extraction, followed by colourimetric analysis (Method 7C2, Rayment and Lyons 2011) for NH_4^+-N and $NO_3^-N + NO_2^-$-N. These values were summed to give a total mineral-N concentration of the material. The mineral-N sum less mineral-N concentration changes in the three background site-deposition traps was used to calculate NH_3-N deposition for the deployment period.

Statistical analyses

Summary statistics were prepared using Student's *t*-tests and ANOVA (all applied using R; R Development Core Team 2014). Probability distributions of deposition-trap data were compared with probability distributions of background deposition-trap data using the fitdist function of the fitdistrplus package in R, and the Kolmogorov–Smirnov test was applied to determine whether sample distributions differed significantly.

Results and discussion

During the measurement period, the dominant wind direction at the site was from east south-east (Fig. 1). Mean wind speeds were ~1.6 m/s, and rainfall during the period was 204 mm. The average temperature for the study period was 24.0°C, close to the average annual temperature (25°C), and the average temperature difference between the manure and air was 5°C.

Measurements of NH_3 were conducted at the background site well removed from the feedlot (1.8 km from the feedlot) and where wind directions did not originate from the feedlot. The median background NH_3 concentrations were ~0.007 mL NH_3 m^3 of air, which were within the range reported previously (0.004–0.010 mL NH_3 m^3 of air; Denmead *et al.* 2008), and <1% of the intercepted air concentrations at the air intake at the feedlot (0.240–4.50 mL NH_3 m^3 of air). With this contrast between the background and feedlot NH_3 concentrations, there

is little risk of error in background determination leading to significant errors in emission estimates.

Ammonia volatilisation

Total NH_3 volatilised from the operation during the period amounted to 210 tonnes of NH_3-N during the study period (Table 1). It is apparent that a higher proportion of volatilisation than suggested by the inventory protocols may be appropriate in this case. This has also been the observation of other studies conducted on this subject (Denmead *et al.* 2008; Loh *et al.* 2008).

The inventory calculations assume that only 30% of excreted N becomes volatilised, using a 430-kg average animal (as indicated by feedlot production statistics), and on the basis of their emission factors (68 g per animal per day for feedlot beef cattle, Intergovernmental Panel on Climate Change 2006; or 65 g per animal per day, Department of Environment 2014). Mean measured volatilisation during the 129-day period was equivalent to 127 g per animal per day (mean of half-hourly measurements; lower and upper 95% confidence limits were 16 and 289 g per animal per day). This value differs somewhat from those of the 2-week long survey in Queensland under warmer conditions (253 g per animal per day) by Loh *et al.* (2008) , but is greater than measurements from the same site for a 2-week winter period (46 g per animal per day, Denmead *et al.* 2008).

Measured volatilisation was about twice the inventory estimates (Department of Environment 2014; Intergovernmental Panel on Climate Change 2006; Table 1). Assuming that the two inventory protocols were otherwise accurate, our measurements suggested that a volatilisation factor of 60%, rather than 30%, may be more appropriate for calculations in this case.

As expected, a diurnal pattern of volatilisation was evident (Fig. 2*a*; Bai *et al.* 2015), and a correlation relationship between wind-friction velocity and NH_3 volatilisation was observed (linear regression $R^2 = 0.23$; Kendall's tau = 0.36; $P < 0.05$; Table 2).

Temperature appears to be a significant driver of NH_3 volatilisation. Significant correlations were observed between total NH_3 volatilisation from the feedlot and manure temperature (measured at 5-mm depth; linear regression $R^2 = 0.2$; Kendall's tau = 0.36; $P < 0.05$; Table 2). This suggests minor temperature-controlled effects on the equilibrium between NH_4^+ and dissolved NH_3 in the manure, as well as the equilibrium between dissolved and gaseous NH_3. A relationship between total NH_3 volatilisation per second from the feedlot and air temperature was also supported by a weak significant correlation between these factors (linear regression $R^2 = 0.06$; Kendall's tau = 0.16; $P < 0.05$; Fig. 2*b*, Table 2). Other researchers have found that NH_3 volatilisation is sensitive to air-temperature relationships by using the DeNitrification–DeComposition model (DNDC) to represent beef feedlot volatilisation data (Waldrip *et al.* 2013).

A stronger relationship was observed by comparing the difference in temperature between manure (at 5-mm depth) and air (driven by solar radiation; consistent with Sommer and Olesen 2000) and NH_3 volatilisation (linear regression $R^2 = 0.38$; Kendall's tau = 0.46; $P < 0.05$; Fig. 2*c*, Table 2). This relationship is consistent with the temperature effect on the Richardson number and stability correction factor (Sommer *et al.* 2003). This highlights the important role of buoyancy-driven convection on NH_3 volatilisation, which is dependent on temperature gradients at the manure surface.

None of the other correlation relationships investigated with volatilisation of NH_3 were significant (cattle numbers, modelled manure moisture, or daily change in manure moisture). Cattle numbers in the immediate pen were significantly, but weakly correlated with measured air concentration of NH_3 (Kendall's tau = 0.21; $P < 0.001$; Table 2). However, atmospheric transport factors have effectively obscured the relationship between emissions and cattle numbers.

The non-linear model of Sommer and Olesen (2000; also Sommer *et al.* 2003), which utilises this temperature-difference relationship, may be applicable in generalising from one site to another for the effect of temperature on NH_3 emission where the environmental and physical factors may fall outside of those at this site. This could be the case, for example, where wind speed, friction velocity and other factors determine the slope and shape of the temperature-difference relationship with emission.

The feedlot layout used in bLs modelling (using Windtrax; Crenna *et al.* 2008) is very similar to that applied by other authors using a single sample intake (Denmead *et al.* 2008). When using the bLs technique with this layout, more distant

Table 1. Cumulative mineral-N data, both for deposition traps and total volatilisation from the feedlot (both measured and two inventory calculations)

The following two inventory protocols were applied and volatilisation estimates (N intake to volatilisation) are included for comparison: the Australia inventory estimate (Department of Environment 2014), and the IPCC estimate (Intergovernmental Panel on Climate Change 2006; 430-kg average cattle weight was used). The deposition entries represent an estimate of total deposition if the pattern represented by the transect was rotated around the entire boundary of the pen area

Parameter	Flux μg ($N/s.m^2$)	95% confidence interval (μg $N/s.m^2$)	Cumulate (tonnes)	Proportion of measured (%)
Measured volatilisation	83.1	21.72–238	210	–
Australian inventory (Department of Environment 2014)	–	–	111	53
IPCC inventory (Intergovernmental Panel on Climate Change 2006)	–	–	105	50
Net deposition, western site	0.25	0.003–2.00	6.7	3.2
Net deposition, southern site	0.211	0.009–1.19	3.6	1.7

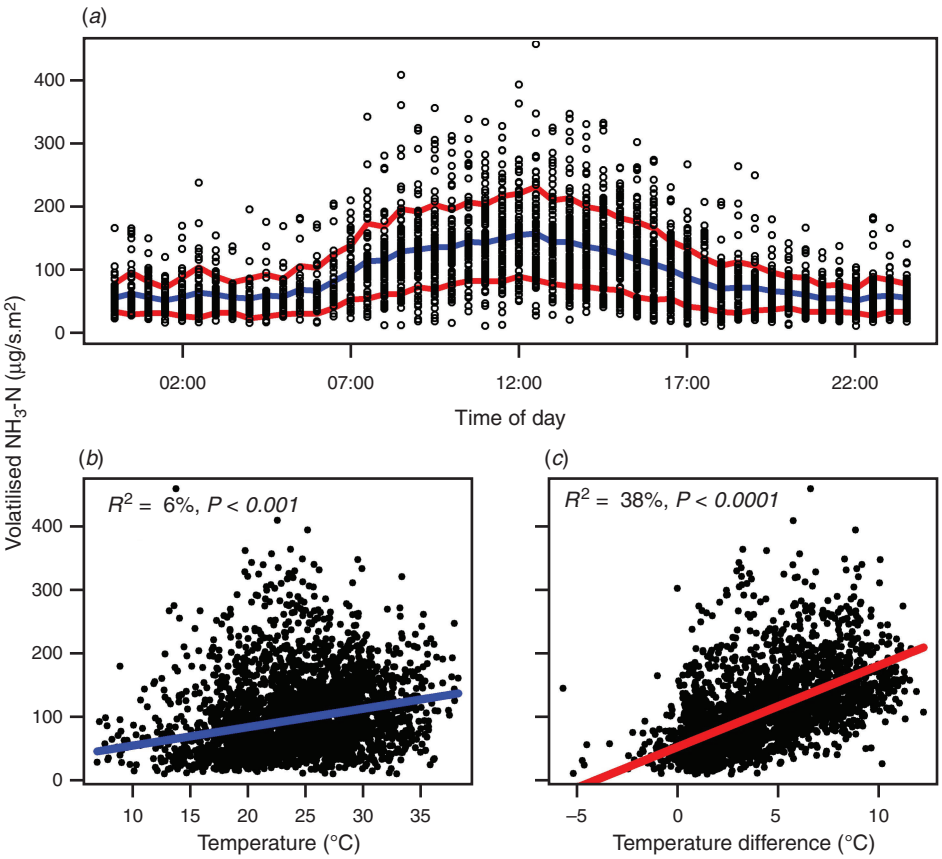

Fig. 2. Ammonia (NH_3) volatilisation flux is related to temperature: (*a*) daily variation in volatilisation; the central line represents the mean, while the two outer lines represent the mean ± standard deviation; (*b*) NH_3 volatilisation versus air temperature, with a linear regression fitted; (*c*) the effect of the difference in temperature between manure (at 5 mm depth) and air on NH_3 emission (plotted as [temperature manure – temperature air] versus NH_3 emission; $R^2 = 0.38$; regression line plotted).

Table 2. Weak to moderate correlation relationships observed between ammonia (NH_3) volatilisation and measured parameters, and between net deposition and measured NH_3 volatilisation

Relationship	Kendall's tau
Wind friction velocity	0.34; $P < 0.001$
Manure temperature	0.36; $P < 0.05$
Air temperature	0.16; $P < 0.05$; Fig. 2*b*
Temperature difference between the manure and air	0.46; $P < 0.05$; Fig. 2*c*
Cattle numbers in pen	0.21; $P < 0.001$
Relationships net deposition and measured volatilisation	0.43; $P = 0.24$

emission sources contribute far less to the emission estimate than do the sources immediately adjacent to the sample intake. As noted previously (Denmead *et al.* 2008), while there were ponds and manure piles at the western outer bound of the pen area, these distant and dominantly down-wind sources have little influence on the measured emissions, as few of the simulated touchdowns were within these regions. Re-running the Windtrax model with a layout that included the pond area as part of the emission source had no significant ($P > 0.05$) effect on the emission flux.

Net nitrogen deposition

Cumulative net deposition (deposition less re-volatilisation) amounts to 6.7 and 3.6 tonnes N along the western and southern transects (respectively), with deposition approaching background rates at the maximum distances from the pens (deposition not significantly greater than background, $P > 0.20$; Figs 3, 4; maximum distance from pens ~600 m). At the approximate midpoints of the deposition-trap transects (251 m for southern and 314 for western), net deposition across all deployments tended to be greater than background ($P = 0.065$). These results are also consistent with recent airborne measurements that indicated a rapid dilution of plume NH_3 concentrations as distance increased from the feedlot (0.470 mL NH_3 m^3 of air at 500 m to 0.022 mL NH_3 m^3 of air at 5100 m; 35 m above ground level; Hacker *et al.* 2016).

There was a strong contrast for each of the deployment periods between the soil concentrations of the more distant traps (background, western 601 m, southern 518 m; 42.8 ± 22.8 mg mineral N per kg of soil) and closer traps ($P < 0.001$; from background to 766 mg mineral N per kg of soil; median western:

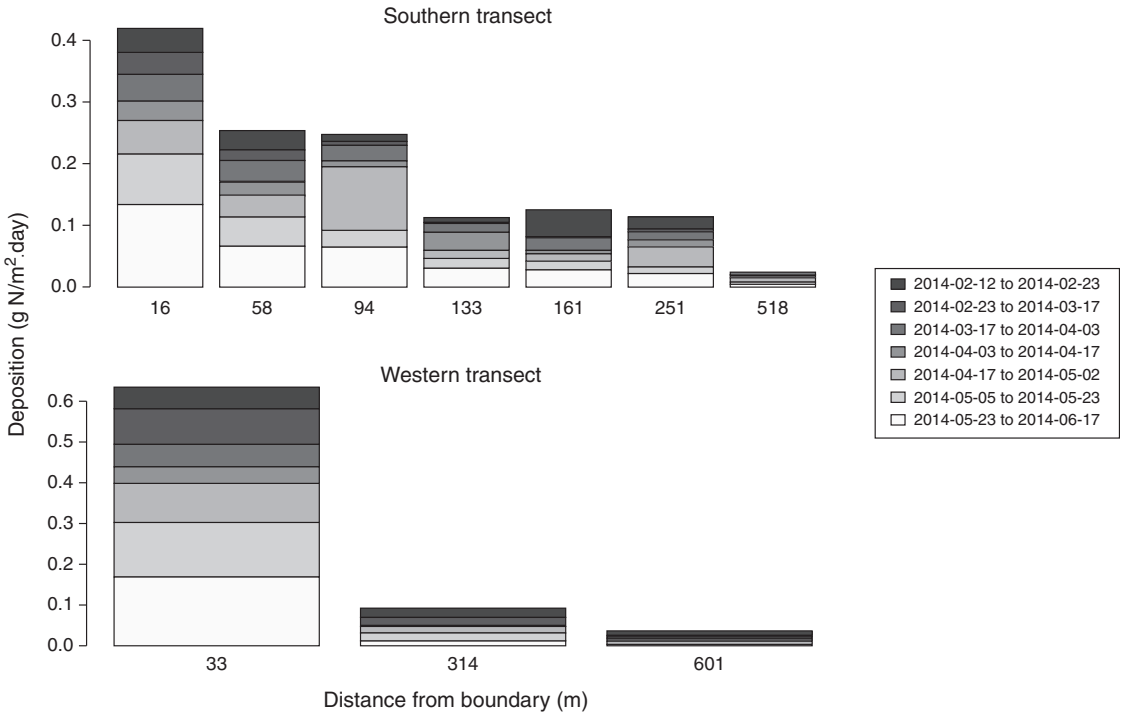

Fig. 3. Net deposition along two transects radiating from the feedlot boundary.

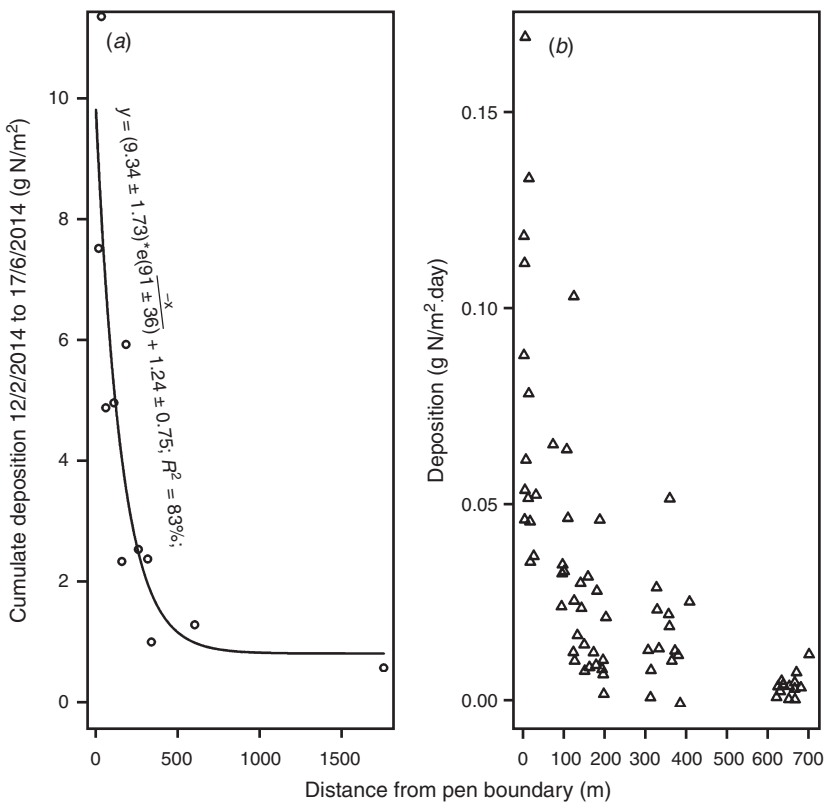

Fig. 4. (*a*) Fitting an exponential decay equation to the cumulative net deposition data suggests that deposition has effectively returned to background levels within 600 m of the feedlot boundary. (*b*) Individual measured ammonia (NH_3) deposition periods as a function of mean distance from the pen boundary.

33 m sample, 196 mg mineral N per kg of soil; median southern: 0 m, 121 mg mineral N per kg of soil).

There was a moderate but non-significant correlation between net deposition during the seven measurement time periods (summing all traps), and the measured volatilisation of the corresponding period (Kendall's tau = 0.43, $P = 0.24$; Table 2). This weak relationship is probably to be expected where important factors such as wind directions are not accounted for in this calculation. However, implementation of a deposition model relating NH_3 deposition at a point to the measured volatilisation, wind direction and turbulence characteristics, distance from the pen boundary, and manure and air temperatures (e.g. Asman 1998) may be more successful.

Layout of the deposition-trap transects was strongly dictated by feedlot infrastructure, management of the feedlot, and cropping of the surrounding areas. The western deposition transect was well aligned with the dominant wind direction (Fig. 1); however, only three locations were available along this transect (the transect consisted of only 3 traps; available sites defined by farm management). The southern transect was representative of a minor wind direction.

On this basis, it is likely that the western transect provides an upper estimate of net deposition, with evidence of ~3% of volatilised NH_3 being deposited within 601 m of the feedlot boundary. The lower limit suggested by the southern transect is ~1.7% of volatilised NH_3 being deposited within 518 m of the pen boundary.

The background site delivered consistently low median air concentrations relative to the pen air concentration (Fig. 1; irrespective of the wind direction, 0.013 mL NH_3 m^3 of air at the background relative to a 0.240–4.500- mL NH_3 m^3 of air range at the pen intake). This suggests that the selected background site appropriately estimates native NH_3 emissions in this landscape, without influence from the feedlot. For additional confidence, mineral-N analyses of the three deposition traps located at the background site were compared with those of the traps at the western (601 m) and southern (518 m) sites, depending on the dominant wind directions during the period. This comparison was conducted to ensure that measured background values were comparable to or lower than those at these sites.

The measured background net deposition of NH_3 plus soil N mineralisation was 0.0167 ± 0.012 g N/m.day (mean ± standard deviation). In reality, almost all of this mineral-N was attributable to soil mineralisation, rather than to background deposition. Analysis of the vials of deposition-trap soil retained in the laboratory at 25°C during each deposition-trap deployment indicated average soil mineralisation of 0.0166 ± 0.0088 g N/m.day (N deposition would therefore be equivalent to ~0.4 kg N/ha.year).

A review of NH_3 deposition rates indicated a range of bulk deposition from 9.2 to 16.8 kg/ha.year, with deposition to plant canopies and grass surfaces of 19.6–95.6 kg/ha.year (Krupa 2003). Such values are likely to be very location specific, and recent data from the United States (http://nadp.sws.uiuc.edu/ntn/annualmapsByYear.aspx) indicate a much more restricted range of ammonium wet deposition, not dissimilar to that estimated for our site (mean total N deposition for ~264 sites, 2012, was 3.2 kg N/ha.year, with 95% confidence interval of 0.2–7.3 kg N/ha.year).

Implications

A detailed investigation of the soil from this site suggests that where mineral-N is <70 mg per kg of soil, no significant N_2O emission occurs (Redding *et al.* 2016). None of the deposition-trap samples collected during 5 months from the southern 518-m site and only one sample collected from western 601-m site exceeded 70 mg mineral-N per kg of soil. However, the depth of soil in the deposition traps (10 mm) may not have realistically represented the depth of interaction of deposited NH_3. In reality, NH_3 may have been restricted to the upper few millimetres of the soil, leading to higher soil concentrations in that shallow zone.

However, several significant observations can be made. First, only a small proportion of the volatilised NH_3 is deposited within 601 m of the feedlot (<3.2%; Table 1). These deposition results are supported by the data of other authors, collected from several locations, where the majority of volatilised N was observed to be advected away (measured <3.2% within 270 m of a poultry farm; Fowler *et al.* 1998; <10% within an 8 × 8 km square; Staebler *et al.* 2009). In this zone, application of an emission factor for indirect emissions (e.g. Intergovernmental Panel on Climate Change 2006) is probably reasonably well supported by evidence. However, soil nutrient-management approaches for efficient plant production may be able to greatly decrease indirect emissions. Maximal recorded mineral-N deposition during the measurement period of 5 months (more than a single crop-growth period) was ~100 kg N/ha, which is similar to a commercial-fertiliser application rate.

Second, beyond 600 m from the pen boundaries, deposition fluxes appear to return to rates that are a small proportion of seasonal crop or pasture requirements (close to background deposition). In our study, deposition in this external zone was probably ~0.4 kg N/ha.year (based on background-site deposition less the soil blank), which may actually not be significantly different from 0 kg N/ha (Fig. 4; fitting an exponential decay curve to the data; 1.24 ± 0.75 g N/m over a 129-day period; effectively not significantly different from 0 g N/m^2). These results are supported by a previous study that found that >500 m from a feedlot, soil N remained at concentrations typical of the surrounding undisturbed shortgrass prairie (Todd *et al.* 2008*b*). However, field measurements at the present study site suggested that background fallow (but cultivated) soil mineral-N concentrations are close to the threshold for N_2O emission (70 mg mineral-N per kg of soil; Redding *et al.* 2016; soil measurements collected in cultivated area adjacent to the background site; Fig. 1; 77 ± 22 mg mineral-N per kg of soil), indicating that deposition in these areas will result in N_2O emission.

The study site was located within a region of intensive grain production, on high-quality agricultural soils. This is the case for many intensive livestock enterprises where grain is an essential feedstock. It is likely that advected mineral-N is deposited to this wider landscape, to soil where mineral-N concentrations are purposely raised through fertiliser applications. This deposition could be considered a manageable fertiliser application with low embodied transport and manufacturing emissions. Where re-deposition coincides with the nutrient uptake of any growing vegetation, these applications are unlikely to remain resident for long (based on sorghum and wheat growth curves estimated via local production data and a logistic curve;

Hunt 1982), meaning that there would be little accumulation potential under these circumstances.

Conclusions

Total NH_3 volatilised from the enterprise during the period amounted to 210 tonnes of NH_3-N during the study period (127 g per animal per day). It is also apparent that the inventory volatilisation factor (30% of excreted N) underestimates volatilisation, in this case, by a factor of two.

For the same period, net deposition within 600 m of the pen boundary is probably between 3.6 and 6.7 tonnes N (1.7–3.2% of volatilised NH_4^+-N), with deposition approaching background rates at the maximum distances of the deposition traps from the pens (601 m for the western transect; 518 m for the southern transect). Background net deposition was measured at ~0.4 kg N/ha.year.

Our study has highlighted the important role of manure–air temperature gradients on emission. The temperature gradient explained ~38% of the variance in the emission rate, whereas a linear model of air temperature on emission explains only 6% of the variance in emission rate.

Conflicts of interest

The authors declare no conflicts of interest.

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