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## Research Paper

# Ammonia emissions from untreated, separated and digested cattle slurry – Effects of slurry type and application strategy on a Swedish clay soil



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## ARTICLE INFO

## Article history:

Received 8 July 2022

Received in revised form

1 December 2022

Accepted 16 January 2023

Published online 28 January 2023

## Keywords:

Trailing hose

Trailing shoe

Slurry acidification

Slurry injection

Slurry separation

ALFAM2

Animal slurry contains plant nutrients such as nitrogen (N) that are essential for crop production. Inorganic slurry N is easily volatilised as ammonia after field application, reducing slurry fertiliser value and causing environmental problems. Ammonia emissions can be reduced by lowering slurry pH, rapid infiltration or incorporation of slurry into soil. This study investigated the effect of different combinations of slurry types and application strategies on ammonia emissions. The slurry types tested were untreated cattle slurry (CS), the liquid fraction from mechanical solid–liquid separation of cattle slurry (LF) and biogas digestate based mainly on cattle slurry (BD). The application strategies tested were trailing hoses, trailing shoes, trailing hose application of acidified slurry and slurry injection. Ammonia emissions after slurry application were measured using wind tunnels, with continuous measurements of ammonia concentrations in outgoing air. Comparisons were also made between measured ammonia emissions and emissions predicted by the ALFAM2 model. Cumulative ammonia emissions after 70 h from LF, CS and BD represented 23%, 29% and 32% of total ammoniacal nitrogen (TAN) applied. Trailing shoes and 50 mm deep injection slots reduced ammonia emissions by on average 17% and 37%, respectively, compared with trailing hoses. Slurry acidification resulted in an average reduction in ammonia emissions of 83%. The ALFAM2 model was reasonably accurate in predicting cumulative emissions (70 h). Accuracy in predicting emission dynamics was low in some cases, likely due to differences between wind tunnel measurements and open-air emissions and to model error.

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<https://doi.org/10.1016/j.biosystemseng.2023.01.012>

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**Nomenclature**

|                    |   |
|--------------------|---|
| A                  | soil area covered by the wind tunnel ( $\text{m}^2$ )               |
| ANOVA              | analysis of variance  |
| BD                 | biogas digestate  |
| C                  | ammonia concentration ( $\text{g N L}^{-1}$ )                       |
| $c_i$              | inlet (background) concentration or mixing ratio ( $\text{ppb}_v$ ) |
| $c_o$              | outlet concentration or mixing ratio ( $\text{ppb}_v$ )             |
| CRDS               | cavity ring-down spectrometer                                       |
| CS                 | untreated cattle slurry   |
| DM                 | dry matter  |
| $F_{\text{NH}_3}$  | ammonia flux ( $\text{g N m}^{-2}$ )                                |
| IHF                | integrated horizontal flux  |
| LF                 | liquid fraction from solid–liquid separation of cattle slurry       |
| M                  | molar mass of nitrogen ( $14.01 \text{ g mol}^{-1}$ )               |
| N                  | nitrogen  |
| $\text{NH}_3$      | ammonia (gas)   |
| $\text{NH}_3$ (aq) | free ammonia in a liquid  |
| $\text{NH}_4^+$    | ammonium  |
| P                  | total pressure (1 atm)  |
| q                  | volumetric airflow rate ( $\text{L min}^{-1}$ )                     |
| R                  | gas constant ( $\text{L atm K}^{-1} \text{ mol}^{-1}$ )             |
| SDG                | sustainable development goals                                       |
| T                  | temperature (K)   |
| TAN                | total ammoniacal nitrogen   |

**1. Introduction****1.1. Background**

Animal manure from livestock production is an important source of nitrogen (N) and other plant nutrients for crop production worldwide (Warren Raffa, Turner, Tubiello, & Serrano, 2018). However, besides being a valuable fertiliser, manure is also an important source of emissions of ammonia ( $\text{NH}_3$ ) and greenhouse gases (Oenema, Bannink, Sommer, Van Groenigen, & Velthof, 2008). Ammonia losses result in lower and more uncertain fertiliser value of the manure when applied to crops. The ammonia emissions from manure are also a health risk for humans and animals, and a cause of environmental problems such as eutrophication and acidification of soils and water (Anderson, Strader, & Davidson, 2003; Webb et al., 2013). In the European Union (EU) and UK, more than 4.8 million tonnes of N are applied to agricultural soils annually, making up about 20% of N input (Misselbrook & Bittman, 2022). In Sweden, one-third of the land area used for crop production receives animal manure, with 84% of the total manure volume applied in liquid form, as slurry (Statistics Sweden, 2020). The most common application technique is trailing hoses, applying 75% of the total slurry volume, followed by broadcast application and slurry injection at 20% and 5%, respectively. With over 56,000 ha in Sweden fertilised with animal slurry yearly (Statistics Sweden, 2020), increased use of ammonia abatement techniques would have positive effects on the environment and on nitrogen use efficiency in crop production.

Ammonia emissions from the agricultural sector and potential subsequent processes, including conversion to nitrate and nitrous oxide, are relevant to several of the United Nations Sustainable Development Goals (SDG), including those regarding human health (SDG 3), clean water (SDG 6), climate change, and life in water and on land (SDG 11–15) (Kanter, Zhang, & Howard, 2016). Ammonia is also one of five pollutants regulated under the EU National Emission Reduction Commitments Directive, with the target of reducing the threat to ecosystem biodiversity and premature deaths caused by air pollutants (European Environment Agency, 2022). Sixteen of the 27 EU member states met their national targets for ammonia reductions in 2020, while the others, including Sweden, need to reduce their emissions further to meet the targets for the period 2020–2029. For these reasons, various approaches are being used to reduce  $\text{NH}_3$  losses from field-applied slurry (Misselbrook & Bittman, 2022).

**1.2. Ammonia emissions and emission abatement**

Ammonia emissions after field application of slurry depend on a number of factors, including soil and slurry properties, application technique and weather conditions such as temperature, solar radiation, wind speed and precipitation (Sommer et al., 2003; Sommer & Hutchings, 2001). The emission rate is highest immediately after slurry application, with 50% of total ammonia losses usually occurring within the first 12 h (Hafner et al., 2018).

Ammonium ( $\text{NH}_4^+$ ) and free ammonia ( $\text{NH}_3$  (aq)) are an acid–base-pair, with the chemical equilibrium between  $\text{NH}_4^+$  and volatile  $\text{NH}_3$  (aq) depending on manure pH. Therefore ammonia emissions are strongly pH-dependent, and slurry acidification can be used for ammonia abatement in animal housing systems, during slurry storage and after field application. The most common additives used to lower the pH are strong acids, e.g. sulphuric acid (Fangueiro, Hjorth, & Gioelli, 2015). In earlier studies testing different acids and with different target pH values, reported ammonia abatement after land application of acidified cattle slurry has been within the range 15–80% (Fangueiro, Hjorth, et al., 2015). In more recent studies, acidification of cattle slurry with sulphuric acid to pH 6.0 or lower has been found to reduce ammonia emissions by on average around 80% (Fangueiro et al., 2017; Pedersen, Feilberg, & Nyord, 2022; Seidel et al., 2017).

Ammonia abatement techniques for slurry application have been investigated and reviewed in a number of publications over the years (Häni, Sintermann, Kupper, Jocher, & Neftel, 2016; Misselbrook, Smith, Johnson, & Pain, 2002; Nicholson, Bhogal, Rollett, Taylor, & Williams, 2018; Seidel et al., 2017; Webb, Pain, Bittman, & Morgan, 2010), with the most common reference technique being broadcast application. The abatement effect achieved by trailing shoes and open-slot injection is generally larger than that from trailing hoses, but the measured effect varies widely for each technique (Webb et al., 2010). In a recent study with trailing hoses as the reference technique, the ammonia abatement from trailing shoes was on average 20%, with the greatest effects seen on more coarse-textured soil (Pedersen, Feilberg, Kamp, Hafner, & Nyord, 2020). Slurry separation, decreasing the slurry dry matter (DM) content of the liquid fraction, has the

potential to significantly reduce ammonia losses after land application of the liquid fraction (Amon, Kryvoruchko, Amon, & Zechmeister-Boltenstern, 2006; Nyord, Hansen, & Birkmose, 2012), presumably as a result of increased slurry infiltration. Studies investigating the effect of slurry injection have reported variable results, with the ammonia abatement effects strongly related to the performance of injectors and the volume of injection slots (Hansen, Sommer, & Madsen, 2003; Rodhe & Etana, 2005).

### 1.3. Measuring ammonia emissions

The techniques available for measuring ammonia emissions in field experiments can be divided into two main categories: micrometeorological methods, requiring large experimental areas, and enclosure methods using smaller experimental plots (Misselbrook, Nicholson, Chambers, & Johnson, 2005). Micrometeorological methods give reliable measurements in terms of absolute ammonia emissions under ambient conditions. Wind tunnels generally use constant airflow, and mass transfer properties differ from open-air conditions and depend on wind tunnel design (Saha, Wu, Zhang, & Bjerg, 2011; Scotto di Perta et al., 2019), and therefore wind tunnel measurements may under- or overestimate emissions (Sommer & Misselbrook, 2016). A new technique combining wind tunnels with continuous online measurements of ammonia concentrations has recently been developed (Pedersen et al., 2020). It provides high-time resolution data, making it possible to monitor emission patterns closely. In addition, the low measurement variability makes it a suitable technique for comparing different low-emission application strategies.

### 1.4. Modelling ammonia emissions

Models for prediction of ammonia emissions can be used for different purposes, e.g. evaluation of mitigation strategies, as a basis for regulations related to field application of animal manure, or for national emissions inventories (Hafner et al., 2019). Several models have been developed during recent decades, some mainly process-based and others of a more empirical nature, with different degrees of complexity and different input requirements (e.g. Congreves et al. (2016); Genermont and Cellier (1997); Nicholson et al. (2013)). Empirical models have the advantage that they are usually less complex, need relatively few input variables and are easy to use. Process-based models, on the other hand, may be more accurate under new conditions and may be better at predicting complex responses (Hafner et al., 2019).

### 1.5. Aim and hypotheses

The aim of this study was to quantify interactive effects between treatments reducing slurry DM content and low-emission slurry application strategies. Ammonia emissions were measured in field experiments with slurry application to grass ley, on a heavy clay soil with 43% clay content. The slurry types used were untreated cattle slurry (CS), the liquid fraction from screw-press separated cattle slurry (LF) and biogas digestate based mainly on cattle slurry (BD). The application strategies used were trailing hoses, trailing shoes,

acidified slurry applied by trailing hose and slurry injection (see Fig. S1 for more information on application methods). The system used, combining wind tunnels with online ammonia measurements, yielded values with high time resolution and low variability, thereby providing accurate detailed data on the ammonia flux patterns throughout the experiments. Flux data from the field experiments were compared against emissions predicted by the ALFAM2 model (Hafner & Haeni, 2022), in order to explore the applicability of the model to wind tunnel measurements and to evaluate its performance and possibly identify areas of interest for further model development. This is especially relevant for acidified slurry and digested slurry, for which the underlying data in the ALFAM2 database are limited.

The hypotheses tested were that ammonia emissions after land application of cattle slurry can be reduced by 1) lowering the slurry DM content by solid–liquid separation or biogas digestion, thereby increasing infiltration, and 2) lowering the slurry pH by slurry acidification. Two further hypotheses tested were that ammonia emissions can be reduced by 3) reducing the slurry area after application and aiding slurry infiltration by trailing shoe application, and 4) further increasing the slurry-soil contact and lowering the exposed surface area by slurry injection. Differences in the effect depending on how these measures were combined were also investigated. The hypotheses in that context were that 5) slurry acidification has the largest effect on digestate, due to higher initial pH and 6) there is no difference in effect between different slurry types when using trailing shoes.

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## 2. Materials and methods

### 2.1. Ammonia emissions experiments

In total, five ammonia emissions experiments were conducted at Lanna field research station in south-west Sweden (58°20'N, 13°7'E) during June and early July 2019. The experiments lasted from 70 h to 120 h, with the shortest time (i.e. 70 h) being selected for comparison of cumulative ammonia emissions. A randomised block design was used in all experiments. In experiments 1–3, application with trailing hoses was compared with application with trailing shoes (Zunhammer GmbH, Traunreut, Germany), together with slurry acidification (with trailing hose application) for each of the three slurry types. Experiment 4 involved comparison of the three slurry types, all applied with trailing hoses. In experiment 5, separated slurry (LF) was used and trailing hose application was compared with simulated slurry injection at two depths, 20 mm and 50 mm (Injection1 and Injection2, chosen to reflect poor and optimal performance of slurry injection, respectively). For both Injection1 and Injection2, a pickaxe (Fig. S1) was used to break the soil surface crust and create the slots, resulting in a 40–50 mm wide band with loosened soil in the uppermost soil layer.

#### 2.1.1. Soil, crop and weather conditions

All experiments were conducted in the same field, on a silty clay soil with 43% clay, 14% sand, 40% silt, 2.6% organic matter and pH 7.0 (1:5 soil:H<sub>2</sub>O). Clay content was analysed using a

sedimentation method modified from Gee and Bauder (1986), the sand fraction was determined by sieving and the residual sample mass was assumed to be the silt fraction. Preceding crop was winter wheat in both 2018 and 2017. In experiments 1–4, the crop was a forage grass ley consisting of 40% timothy (*Phleum pratense* L.), 30% meadow fescue (*Festuca pratensis* L.) and 30% perennial ryegrass (*Lolium perenne* L.), sown in September 2018 and with a first cut on 27 May 2019, before experiment start. Experiment 5 was conducted in an adjacent part of the same field, in spring barley stubble. The barley (flowering completed, crop still green) was cut with a grass plot harvester three days before experiment start and removed from the field. It was expected that spring cultivation would result in a less compact uppermost soil layer and thereby more optimal conditions for ammonia emissions abatement from slurry injection. Soil water content and soil dry bulk density in the top 50 mm were determined by collection of soil cores ( $n = 4$ ) at 0–50 mm depth in grass ley before the start of experiment 1 and in barley stubble after the end of experiment 5. The soil was first dried at 55 °C for >48 h, followed by 6 h at 105 °C. Soil dry bulk density was 1.05 g mm<sup>-3</sup> (sd = 0.06) in grass ley and 1.07 g mm<sup>-3</sup> (sd = 0.02) in barley stubble. Soil water content was 0.21 g g<sup>-1</sup> (sd = 0.015) at the start of the experimental period and 0.15 g g<sup>-1</sup> (sd = 0.016) at the end. Weather data were obtained from a weather station at the experimental site. Start times and temperature data for the different experiments are summarised in Table 1.

### 2.1.2. Slurry types and application

The untreated (CS) and separated (LF) slurry were obtained from the same commercial organic dairy farm. The untreated slurry was collected from a pit outside the cowshed, from where slurry is pumped to a screw press separator (CRI-MAN SM 260/75 FA DM, CRI-MAN, Correggio, Italy), removing approximately 50% of the DM, creating a solid fraction used as bedding material for the cows and a liquid fraction that is stored in a large storage tank until field application. The separated slurry for the experiments was collected from the storage tank. The biogas digestate (BD) came from a biogas plant processing cattle slurry from about 20 different farms, together with other substrates such as pig slurry and waste products from slaughterhouses and the food industry. The amount of cattle slurry in the substrate mix was about 65% (fresh mass basis). All slurry types were collected at the beginning of April and stored in 1000-L plastic tanks in a barn until experiment start.

Shortly after slurry collection, one representative sample of each slurry type was sent for laboratory analysis, the results of which are shown in Table S1. Slurries were analysed for total ammoniacal nitrogen (TAN), total carbon (C), nitrogen (N), calcium (Ca), potassium (K), magnesium (Mg), sodium (Na), phosphorus (P), sulphur (S) and pH. TAN was extracted with 2 M KCl according to modified Swedish standard ISO 11732 (International Organization for Standardization [ISO], 1995) and analysed using a flow injection analyser (FIStar Analyzer (5000), FOSS, Hilleroed, Denmark). Total C and N were analysed according to modified standards ISO 10694 (ISO, 2005) and ISO 13878 (ISO, 1998), respectively, with combustion of dried slurry samples followed by elemental analysis (928 Series Macro Determinator (model CN928), LECO, St. Joseph, MI, USA). Total Ca, K, Mg, Na, P and S were determined according to modified Swedish standard SS 028311 (ISO, 2017) by extraction with 7 M HNO<sub>3</sub>, followed by analysis of the elements using an ICP spectrometer (SPECTROBLUE ICP-OES, SPECTRO Analytical Instruments GmbH, Kleve, Germany). At the laboratory, slurry pH was measured at room temperature with a glass pH electrode (Jenway™ 924005 pH Temperature Electrode, Cole-Parmer, Stone, Staffordshire, UK). Slurry pH at acidification and field application was measured at ambient temperature with a pH-meter (MW102, Milwaukee Instruments Kft., Szeged, Hungary).

The TAN content was used to calculate slurry application rates corresponding to 60 kg ha<sup>-1</sup> TAN for the different treatments. This N rate was chosen as it is within the range for normal summer N application to grass ley, and the resulting amounts of slurry applied in the experiments were 35.0 metric tonnes ha<sup>-1</sup> for CS and LF, and 17.5 metric tonnes ha<sup>-1</sup> for BD. At the start of each experiment, a new slurry sample was taken (in experiment 4, one sample for each slurry type) and sent for laboratory analysis of DM content and TAN (Table 2).

For each of the experiments with slurry acidification, 5 L of slurry were acidified by manual addition of concentrated (96%) sulphuric acid while mixing until the target pH was reached. For practical reasons, and to give time for the foam that formed to wane, acidification was performed 2–4 h before slurry application. The target pH was set to 6.0 for CS and LF, as a balance between maximising ammonia abatement and minimising the amount of acid added. For BD the target pH was 6.7, to match the pH value in a concurrent fertilisation field experiment using the same slurry types, where the acidification of BD to pH 6.0 failed due to high buffering capacity and extensive foaming.

**Table 1 – Start time, duration and temperature data for ammonia emissions experiments 1–5. Values within brackets represent minimum and maximum temperature.**

| Experiment | Experiment start<br>(yyyy-mm-dd and time) | Experiment duration<br>(h) | Air temp. during<br>slurry application<br>(°C) | Air temp., mean, 70 h<br>(°C) |
|------------|---|----------------------------|--|-------------------------------|
| 1          | 2019-06-05 10:30                          | 73                         | 25.6   | 19.3 (7.6, 28.9)              |
| 2          | 2019-06-13, 12:30                         | 94                         | 21.8   | 18.2 (8.7, 25.3)              |
| 3          | 2019-06-18, 13:00                         | 122                        | 24.6   | 18.3 (10.9, 25.8)             |
| 4          | 2019-06-25, 11:00                         | 71                         | 22.1   | 19.2 (9.3, 27.5)              |
| 5          | 2019-07-02, 13:45                         | 93                         | 19.7   | 14.9 (6.0, 26.1)              |

**Table 2 – Overview of slurry types, slurry characteristics and application rates used in experiments 1–5.**

| Experiment | Slurry type    | DM<br>(%) | TAN<br>(kg ton <sup>-1</sup> ) | Application rate<br>(kg TAN ha <sup>-1</sup> ) | pH               | H <sub>2</sub> SO <sub>4</sub> (96%) added<br>(kg ton <sup>-1</sup> ) | pH acidified |
|------------|----------------|-----------|--------------------------------|--|------------------|---|--------------|
| 1          | Separated (LF) | 4.8       | 1.8                            | 64   | 7.1 <sup>a</sup> | 7.4   | 6.0          |
| 2          | Untreated (CS) | 9.5       | 1.9                            | 65   | 6.8 <sup>a</sup> | 8.3   | 6.0          |
| 3          | Digested (BD)  | 3.9       | 3.2                            | 56   | 7.6 <sup>a</sup> | 11.0  | 6.7          |
| 4          | Untreated (CS) | 9.0       | 1.9                            | 66   | 6.8              | 0   | –            |
| 4          | Separated (LF) | 4.6       | 2.0                            | 71   | 7.1              | 0   | –            |
| 4          | Digested (BD)  | 4.4       | 3.4                            | 60   | 7.8              | 0   | –            |
| 5          | Separated (LF) | 4.5       | 2.0                            | 72   | 7.1 <sup>a</sup> | 0   | –            |

<sup>a</sup> pH measured on different samples from the same slurry types on other dates (June 25 for CS and LF, June 14 for BD).

Before the start of each experiment, the grass height within the plot area in experiments 1–4 was manually adjusted with scissors to 50–70 mm, to make it similar in all experiments. For slurry application, a 10-L watering can equipped with a watering hose was used to mimic trailing hose application. The slurry was applied in three bands at 0.25 m spacing within a metal frame (293 mm × 674 mm inner dimensions), inserted to 40 mm depth into the soil. Immediately after slurry application, the wind tunnel was mounted upon the frame and sealed to be airtight. In treatments with slurry injection or trailing shoe application, the soil slots were created before mounting the frame. Trailing shoe soil slots were created manually one by one with a trailing shoe disassembled from a Zunhammer slurry spreader, with the aim of mimicking those made by the slurry spreader in fertilisation experiments on the same experimental site. Injection slots were created manually with a two-sided metal pickaxe. One of the 90° corners of the broad (axe) end was used to create the 20 mm deep injection soil slots (Injection1), and the pointed (pick) side was used for the 50 mm deep (Injection2) slots (Fig. S1).

### 2.1.3. Wind tunnels and ammonia emissions measurements

The technique of using wind tunnels for ammonia emissions experiments has been described previously (e.g. by Lockyer (1984) and Misselbrook et al. (2005)). Full details of the experimental equipment and procedures for online ammonia measurements used in the experiments can be found in Pedersen et al. (2020). The setup comprised nine wind tunnels, which allowed for three treatments with three replicates per experiment. Each wind tunnel consisted of a rectangular, open-bottomed stainless steel emissions chamber connected to a fan via a steel duct. A small (335 mm × 13 mm) air inlet was positioned in the end of the wind tunnel, and the fan created a constant airflow through the tunnel (air exchange rate 25 min<sup>-1</sup>, resulting in a calculated mean air velocity of 0.33 m s<sup>-1</sup>). From the wind tunnels, air was led through insulated Teflon tubes heated to approximately 40 °C, via a valve block controlling which tube to measure from, to a cavity ring-down spectrometer (CRDS) (G2103 NH<sub>3</sub> Concentration Analyzer, Picarro, CA, USA). Measurement interval of the instrument was 1 s and the measurement units were ppb, with a lower instrument detection limit of 0.03 ppb. Background ammonia emissions were measured at the air inlet of three of the tunnels, and one overall background measurement was made about 20 m

away from the experimental site. The concentration of ammonia in the air from one measuring point was measured for 8 min, which was sufficient for reaching stable values under summer conditions. With 13 measuring points in total (9 wind tunnels, 4 background measurements), a complete round of measurements took 104 min. Data from the CRDS included measurements of ammonia concentration every second, as well as 30-s averages. The last 30-s average from each 8-min measuring period was used for ammonia flux calculations.

### 2.1.4. Data treatment and statistical analysis

For data treatment and statistical analysis, R (version 4.1.2) was used (R Core Team, 2022). The ammonia concentration data from the CRDS were converted from ppb to units of g N L<sup>-1</sup> and corrected for background concentrations based on the ideal gas law:

$$C = M \cdot (c_o - c_i) \cdot P / (R \cdot T) \quad (1)$$

where C is ammonia concentration (g N L<sup>-1</sup>), P is total pressure (1 atm), R is the gas constant (0.08206 L atm K<sup>-1</sup> mol<sup>-1</sup>), T is temperature (K), c<sub>o</sub> is outlet concentration or mixing ratio (ppb<sub>v</sub>), c<sub>i</sub> is inlet (background) concentration (ppb<sub>v</sub>) and M is molar mass of nitrogen (14.01 g mol<sup>-1</sup>) (equation (1) modified from Pedersen et al. (2020)). When outlet concentration was not higher than inlet concentration, C was set to zero. From the ammonia concentration calculated by equation (1), volatilisation flux of ammonia, in g N m<sup>-2</sup>, was calculated as:

$$F_{\text{NH}_3} = C \cdot q / A \quad (2)$$

where F<sub>NH<sub>3</sub></sub> is ammonia flux (g N m<sup>-2</sup>), C is ammonia concentration (g L<sup>-1</sup>), q is volumetric airflow rate (2016 L min<sup>-1</sup>) and A is area under the wind tunnel (0.2 m<sup>2</sup>) (Pedersen et al., 2020).

Cumulative ammonia emissions were calculated using the trapezoidal rule (Burden & Faires, 2001). Data gaps occurred in experiments 1, 2 and 3 (in experiment 1 at 45.1–50.7 h from start, in experiment 2 at 17.3–22.5 h from start and in experiment 3 at 29.5–33.7 h from start). Ammonia flux within the data gaps was estimated using linear interpolation between the two data points on each side of the data gap. In experiment 2, vacuum in the tubing system between the wind tunnels and the CRDS resulted in very low and unreliable data during the first measurement cycle. Correction for the low initial measurements was made by replacing the ammonia concentration values from the first measuring cycle by the values from the following measurement. This assumption

was based on the emission patterns in the other experiments, where the values in the first measurement cycle were 76–136% of those at the next measurement. The difference in total emissions after 70 h before and after this correction was less than 4% for all treatments.

Statistical analysis of the results from the wind tunnel experiments was performed for each experiment separately. Analysis of variance (ANOVA) was carried out using a linear mixed-effects model, `lmer()` function, in the R package `lme4` (Bates, Maechler, Bolker, & Walker, 2015). For the ANOVA analyses, type III sums of squares and F-tests were used, and degrees of freedom were calculated with the Kenward–Roger method. Normality of the data was checked by normal probability plots, where sample quantiles are plotted against theoretical normal quantiles (Q–Q-plot). Homogeneity of variance within the data was checked by plotting model residuals against fitted values. In addition, the `powerTransform()` function in the R-package `car` (Fox & Weisberg, 2019) was used to determine whether transformation of data with the Box–Cox method (Box & Cox, 1964) would significantly increase model likelihood, and to find the optimal power transformation. For experiment 5, this resulted in transformation of the data to a power of  $-3$  prior to the ANOVA analysis. Thereafter, the estimated means and the 95% confidence interval limits were back-transformed to the original scale.

The response variable in all analyses was cumulative ammonia emissions at 70 h, as fraction of TAN applied. Predictor variables in all single-experiment analyses were treatment and block, with treatment always set as a fixed factor and block always set as random factor. The overall analysis of experiments 1–3 used the same predictor variables, with the addition of experiment as a fixed variable. The R package `emmeans` (Lenth et al., 2022) was used for estimation of least-squares means, contrasts and related confidence intervals and *p*-values. Pairwise differences were analysed with the Tukey method and a significance level of 0.05.

## 2.2. Modelling ammonia emissions with the ALFAM2 model

The R version of the ALFAM2 model (v2.0) (Hafner & Haeni, 2022) was used in the modelling work in this study. The ALFAM2 model is a semi-empirical dynamic model for prediction of ammonia emissions, described in detail by Hafner et al. (2019). The latest model parameter values (Set 2) are based on measured ammonia emissions from field experiments in six countries and in total over 600 experimental field plots from the ALFAM2 database (Hafner et al., 2018). In the ALFAM2 model, values for most parameters (including those for wind speed) were based on micrometeorological measurements only, and therefore application to wind tunnel measurements is not straightforward (see also Sections 1.4 and 4.2). Parameter Set 2 is described in more detail in Hafner, Nyord, Sommer, and Adamsen (2021).

In the model, ammonia emissions are calculated based on the predictor variables slurry DM, application method, application rate, incorporation (shallow or deep), air temperature, wind speed and rainfall rate. Slurry TAN is divided into a “fast” pool and a “slow” pool from which emissions occur. This

division of slurry TAN between the pools and rates of emission from the pools are quantified by a set of so-called primary parameters. The value of each primary parameter is based on a number of predictor variables, each combined with a coefficient (secondary parameter), the default values of which are determined by fitting the model with emissions data from the ALFAM2 database (Hafner et al., 2021).

For the experiments with wind tunnel measurements, precipitation was set to zero and wind speed was set to the average air velocity inside the wind tunnels, calculated from volumetric airflow rate and cross-sectional area. This substitution of average wind tunnel velocity for average open-air wind speed is undoubtedly imperfect, but it provides a simple way to evaluate the model using wind tunnel measurements (see Section 4.2 for more discussion on this topic). Slurry characteristics and pH values in the different experiments (Table 2) were used in the modelling. By comparing the emissions predicted by the model with those measured in the wind tunnels, it was possible to evaluate model performance, both in terms of flux patterns over time and as cumulative ammonia emissions after 70 h.

## 3. Results

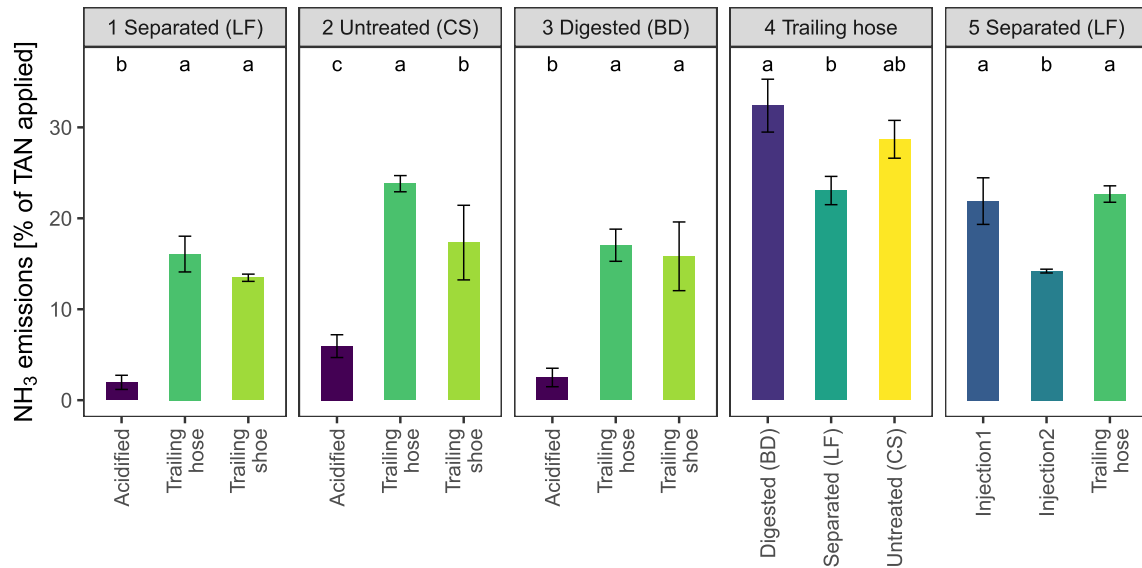
### 3.1. Ammonia emissions

Cumulative ammonia emissions after 70 h varied between 2 and 32% of TAN, depending on the combination of application method and slurry type (Fig. 1). In the experiments with ammonia measurements lasting longer than 70 h (90–120 h), in most cases more than 90% of the total emissions occurred within the first 70 h after application (Table S2). The exceptions were acidified CS in experiment 2, with 79% of total emissions occurring within the first 70 h, and injection of LF to 50 mm in experiment 5, with 89% within the first 70 h, the total measuring time being around 90 h in both experiments (Table S2).

In most cases, 50% of the total ammonia emissions occurred within the first 14 h after slurry application (Table 3). For CS and LF, the time to reach 50% of total emissions was on average 11 h for all experiments with trailing hose application, while for BD with higher initial emissions rate it was only 6 h. In experiment 5, slurry injection slowed down the emissions, with the time taken to reach 50% of total emissions almost doubling for injection to 50 mm compared with trailing hose application (Table 3). The differences between trailing hoses and trailing shoes were small. Slurry acidification had varying effects, with acidified CS and LF taking the longest time of all treatments to reach 50% of total emissions and acidified BD taking the shortest time.

#### 3.1.1. Slurry types

Cumulative ammonia emissions 70 h after trailing hose application of LF, CS and BD in experiment 4 represented 23%, 29% and 32% of applied TAN, respectively (Table 3). For LF and BD, the differences relative to CS were non-significant at 0.05 level ( $p = 0.078$  and  $p = 0.224$ , respectively.) The highest initial emission rate was seen in BD, while CS showed the slowest decline in emissions (Fig. 2).



**Fig. 1 – Cumulative ammonia emissions 70 h after slurry application (n = 3). Error bars represent 1 sd. Within each experiment (1–5), treatments with different lowercase letters are significantly different from each other.**

### 3.1.2. Application strategies

The reduction in ammonia emissions for slurry application by trailing shoe compared with trailing hose was largest for CS (27%,  $p = 0.036$ ), while it was lower and non-significant for LF (16%,  $p = 0.127$ ) and BD (7%,  $p = 0.826$ ) (Table 3).

For slurry injection in 50 mm deep open slots, cumulative ammonia emissions after 70 h were reduced by 37% ( $p < 0.001$ ) compared with trailing hoses, while no difference was seen between trailing hoses and 20 mm injection slots (Table 3). The deep (50 mm) injection slots reduced ammonia flux

compared with trailing hoses for more than 24 h, while the shallow (20 mm) soil slots did not reduce the emissions (Fig. 2).

### 3.1.3. Slurry acidification

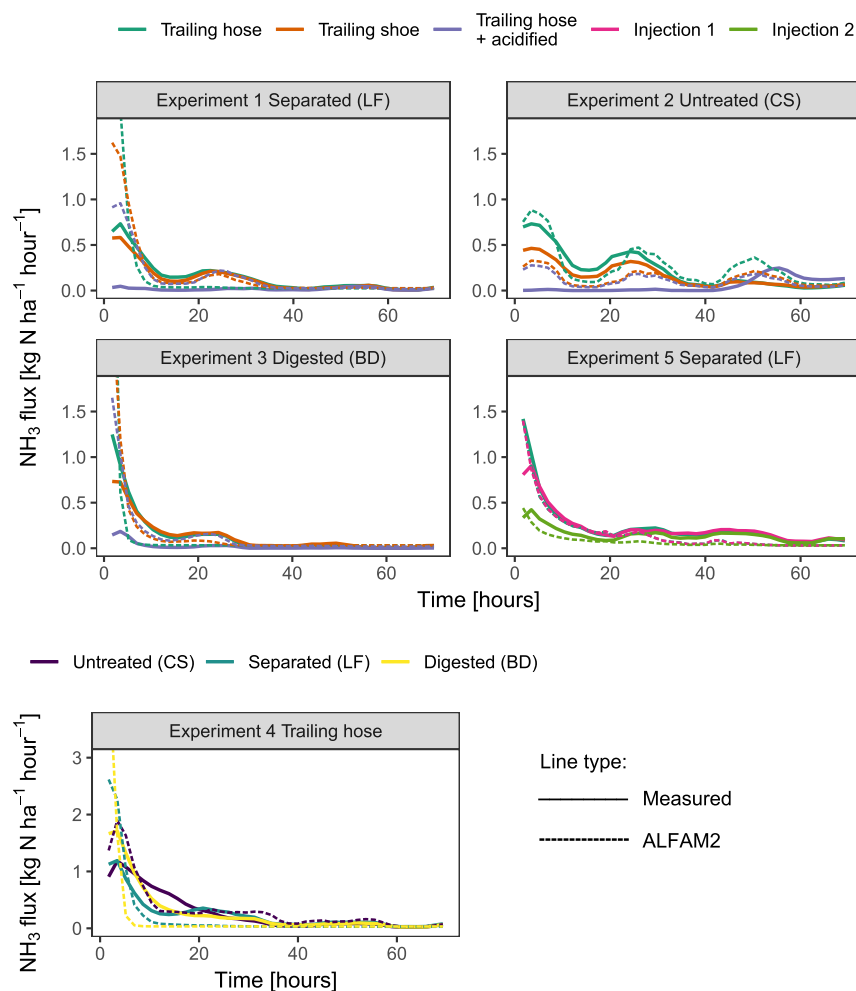
Slurry acidification reduced cumulative ammonia emissions after 70 h for BD by 85% ( $p = 0.004$ ), for LF by 88% ( $p < 0.00103$ ) and for CS by 75% ( $p = 0.001$ ) (Table 3). The emission patterns after acidification differed between the slurry types (Fig. 2). Acidified BD had the highest initial flux, with over 50% of the total ammonia emissions occurring within 5 h from slurry

**Table 3 – Cumulative ammonia emissions after 70 h from experiment start. Within each experiment, treatments with different lowercase letter are significantly different from each other (letters and p-values based on Tukey's HSD test).**

| Experiment and slurry type | Treatment              | Cumulative NH <sub>3</sub> emissions 70 h |                       |   | Time for 50% of cumulative NH <sub>3</sub> loss |
|----------------------------|------------------------|---|-----------------------|---|---|
|                            |                        | Percent of TAN applied                    | 95% confidence limits | p-value for the difference relative to reference treatment <sup>b</sup> | Hours from experiment start                     |
| 1 Separated slurry (LF)    | Trailing hose          | 16.1 a                                    | 14.3, 17.8            | 0.1272  | 10  |
|                            | Trailing shoe          | 13.5 a                                    | 11.7, 15.2            |   |   |
|                            | Acidified <sup>a</sup> | 2.0 b                                     | 0.2, 3.7              |   |   |
| 2 Untreated slurry (CS)    | Trailing hose          | 23.8 a                                    | 20.0, 27.6            | 0.0362  | 13  |
|                            | Trailing shoe          | 17.3 b                                    | 13.5, 21.2            |   |   |
|                            | Acidified <sup>a</sup> | 5.9 c                                     | 2.1, 9.8              |   |   |
| 3 Digested slurry (BD)     | Trailing hose          | 17.0 a                                    | 13.5, 20.5            | 0.8263  | 5   |
|                            | Trailing shoe          | 15.8 a                                    | 12.3, 19.3            |   |   |
|                            | Acidified <sup>a</sup> | 2.5 b                                     | –1.0, 6.0             |   |   |
| 4 All slurry types         | Untreated <sup>a</sup> | 28.7 ab                                   | 25.5, 31.9            | 0.0784  | 10  |
|                            | Separated <sup>b</sup> | 23.1 b                                    | 19.9, 26.2            |   |   |
|                            | Digested <sup>a</sup>  | 32.4 a                                    | 29.2, 35.6            |   |   |
| 5 Separated slurry (LF)    | Trailing hose          | 22.6 a                                    | 20.5, 25.9            | 0.7118  | 14  |
|                            | Injection1             | 21.5 a                                    | 19.8, 24.1            |   |   |
|                            | Injection2             | 14.2 b                                    | 13.8, 14.6            |   |   |

<sup>a</sup> Slurry applied by trailing hose.

<sup>b</sup> Reference treatment for comparison of slurry types in experiment 4 is Untreated; reference treatment for comparison of application strategies in experiments 1–3 and 5 is Trailing hose.



**Fig. 2 – Ammonia flux over time in experiments 1–5. For experiments 1, 2 and 4, extreme ALFAM2 values (max 4.7 kg N ha<sup>-1</sup> h<sup>-1</sup>) immediately after slurry application have been excluded from the diagrams.**

application (Fig. 2, Table 3), and negligible emissions from 30 h until experiment end (Fig. 2). For CS and LF, the initial flux was low and 50% of total ammonia emissions was reached after 49 and 39 h, respectively (Table 3). For acidified CS, the low initial emissions were followed by an increase after around 48 h, with higher emissions continuing until around 70 h after slurry application (Fig. 2).

### 3.2. Ammonia emissions modelled with ALFAM2

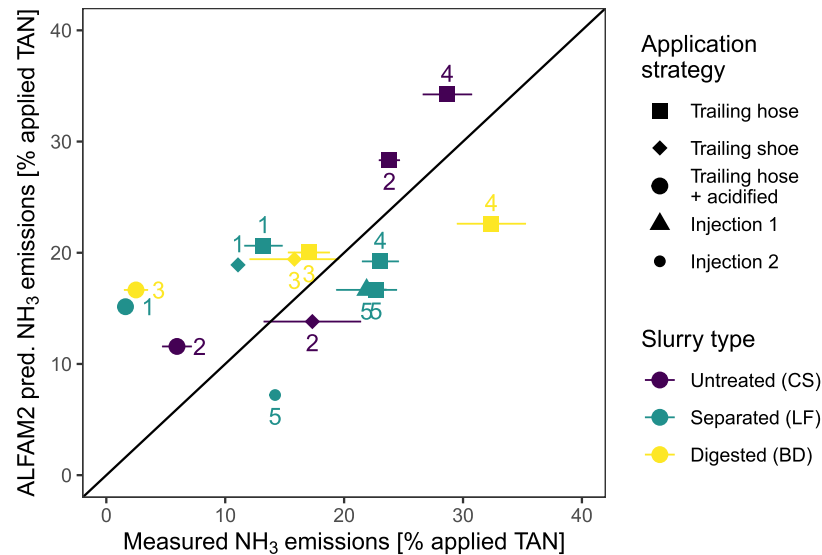
The ALFAM2 model with parameter set 2 predicted cumulative ammonia loss at 70 h with reasonable accuracy across all experiments (Fig. 3). The error ranged from -13 to +15% of applied TAN. Median model absolute error was 6% of applied TAN or 30% of measured emissions.

Differences between trailing hose, trailing shoe, trailing hose with acidification and open slot injection to 50 mm were approximately captured. However, the model substantially underestimated the effect of acidification for LF and BD. Although absolute cumulative emissions were relatively low for measurements (<5%) and for the model (10–20%),

predicted ammonia emissions from acidified slurries were 7-fold and 8-fold higher, respectively, than those measured in wind tunnels. Both model and measurements showed a reduction in emissions from slurry separation in experiment 4, but this agreement did not hold for comparisons across experiments, where other variables presumably had an effect. Anaerobic digestion did not consistently affect measured or model-predicted emissions.

Comparison of measured ammonia flux against model predictions showed varying model performance. In three experiments (1, 3 and 4), the ALFAM2 model predicted much higher flux immediately after application than found in measurements (Fig. 2). In some cases, these high fluxes were followed by under-prediction of later fluxes, e.g., trailing hose application of LF and BD (Fig. 2). Diurnal patterns in measured flux (generally an increase at midday, but with diminishing magnitude over days) were approximately captured by the model in some cases (e.g. trailing hose in experiment 2). For acidified slurry in experiment 2, the overall increase in emission rate after 2 days was not reflected in model predictions.





**Fig. 3** – Measured cumulative ammonia emissions at 70 h from slurry application compared with values predicted by the ALFAM2 model, error bars representing 1 standard deviation. Experiment number is indicated beside each symbol. For observations above the solid black line, ALFAM2 over-predicted emissions compared with measurements, while values below the solid black line are under-predictions.

## 4. Discussion

### 4.1. Ammonia emissions

#### 4.1.1. Effects related to weather parameters

No correlation was found between total ammonia emissions and mean temperature during the experiments or temperature at experiment start (Table 1). This is in line with results from previous modelling work by Pedersen, Nyord, Feilberg, and Labouriau (2021) based on data from wind tunnel experiments, which showed that if air temperature exceeds  $\sim 14$  °C at slurry application, ammonia emissions do not increase with increasing temperature. Balsari, Dinuccio, Santoro, and Gioelli (2008) observed greater ammonia emissions from slurry application under summer conditions compared with autumn application, probably related to a considerably larger temperature span (mean experiment temperature 27.8 °C and 12.5 °C for summer and autumn, respectively) than in the present study.

Rainfall is known to cause crust formation at the surface of clay soils and thereby reduce the infiltration capacity (Lado, Ben-Hur, & Shainberg, 2007; Morin, Benyamini, & Michaeli, 1981). In the present study, a thunderstorm with hail and intense rainfall occurred six days before the start of experiment 4, followed by a period with no precipitation and increasing temperatures. This could possibly have contributed to the greater ammonia emissions during experiment 4 compared with experiments 1–3 (Fig. 1). For slurry types with low DM content in particular (such as LF and BD), the reduced infiltration capacity could have caused increased spreading out of slurry on the soil surface, and thereby greater ammonia emissions. In experiment 5, the greater overall emissions were most likely caused by factors other than crust formation, since

a dense barley crop protected the soil surface during the thunderstorm.

#### 4.1.2. Effects of DM content and slurry pH

Low DM content, as for BD and LF in the present study, may increase the infiltration rate of slurry into the soil and thereby lower ammonia emissions (Bhandral et al., 2009; Pedersen, Nyord, Feilberg, Labouriau, et al., 2021; Sommer, Jensen, Clausen, & Søggaard, 2006). In the present study, this was evident in the faster decline in emission rate for BD and LF compared with CS in experiment 4 (Fig. 2). Pedersen, Andersson, et al. (2021), who used the same slurry types as in our experiments, reported varying results depending on soil texture, with lower ammonia emissions from LF compared with CS in an experiment on clay soil, but higher ammonia emissions from a sandy loam, after application by trailing hoses. Only a few studies have included trailing hose application of the liquid fraction from separated slurry, but e.g. Amon et al. (2006) and Fangueiro, Pereira, et al. (2015), have reported lower total ammonia emissions from the liquid fraction compared with untreated cattle slurry. Studies on broadcast spreading report decreasing (Balsari et al., 2008; Owusu-Twum et al., 2017) or increasing (Dinuccio, Berg et al., 2011) ammonia emissions after application of the liquid fraction compared with the untreated slurry, as well as varying effects (Bhandral et al., 2009; Vandre, Clemens, Goldbach, & Kaupenjohann, 1997). Regarding important parameters such as infiltration, the same behaviour as seen for broadcast spreading can be expected for band application, as there is no soil manipulation with either of those application methods.

The low DM content in biogas digestate is counteracted by high pH, posing a greater risk of ammonia losses. In the present study, the higher initial emission rate from BD compared with LF was most likely a pH-related effect. Other studies, e.g.

Nicholson et al. (2018) and Amon et al. (2006), report greater emissions from biogas digestate than from untreated cattle slurry, while Pedersen, Andersson, et al. (2021) observed higher emissions from biogas digestate on a sandy loam soil, but no difference on clay soil.

The application rate in tonnes per ha was 50% lower for BD. Results from earlier studies on whether that would result in greater ammonia emissions expressed as % of TAN applied are inconsistent. Thompson, Pain, and Rees (1990) found decreasing relative ammonia emissions with increasing application rate, while Balsari et al. (2008) did not observe any difference. Klarenbeek and Bruins (1991) found a non-linear relationship, with reduced relative emissions when application rate increased from 30 to 90 m<sup>3</sup> ha<sup>-1</sup>, but no difference at lower application rates between 10 and 30 m<sup>3</sup> ha<sup>-1</sup>. In parallel experiments to the present study with the same slurry types applied with trailing hoses on clay soil (Pedersen, Andersson, et al., 2021), but with the same volumetric application rate of all slurry types, the differences in relative emissions between slurries, expressed as % of TAN, were very similar to those in the present study.

#### 4.1.3. Effect of application method

4.1.3.1. *Trailing shoes – experiments 1, 2 and 3.* The average ammonia abatement of 17% is in line with findings by Pedersen et al. (2020), but lower than the >50% reduction reported by Malgeryd (1998), although there were large differences between the slurry types. The performance of the trailing shoes was affected by the hard, dry surface of the clay soil, resulting in rather shallow (approx. 10 mm deep) soil slots with very little loose soil, and hence little or no reduction in exposed slurry area compared with application with trailing hoses. On softer soil, the trailing shoe would ideally make approximately 30 mm deep slots, filled up to 10 mm with loose soil, to enable quick slurry infiltration. The study by Pedersen et al. (2020) indicated that the abatement effect of trailing shoes is dependent on soil properties, with the largest effects on more coarse-textured soils.

Larger slurry area has been shown to increase ammonia emissions (Sommer & Hutchings, 2001; Webb et al., 2010). The greater abatement effect for CS (27%) compared with LF (16%) could be attributable to the higher DM content of CS, reducing the spreading out of slurry beside the trailing shoe soil slots. For BD, the lower application rate, leading to narrower slurry bands and quick infiltration for both trailing hose and trailing shoe application, could explain the small ammonia abatement from trailing shoes (7%).

In earlier studies in which trailing hoses and trailing shoes were compared with broadcast application, the abatement effects were in some cases similar, e.g. Häni et al. (2016) found values of 51% and 53%, respectively. In other cases, trailing shoes have been found to be more effective than trailing hoses, e.g. Misselbrook et al. (2002) found a 57% and 26% emissions reduction, respectively. The DM content and pH of the slurry types in those two studies make them most comparable with LF in the present study, where the differences between trailing hoses and trailing shoes were non-significant. Misselbrook et al. (2002) found the weakest abatement effect from trailing shoe application to grass ley in experiments with very short grass and newly established (less

dense) crop, conditions similar to those in experiments 1–4 in the present study.

4.1.3.2. *Slurry injection – experiment 5.* The 50 mm injection slots (Injection2) are most comparable with disc injection, but with slurry applied on the surface rather than injected belowground, while the shallow soil slots (Injection1) are comparable with trailing shoe application. The 37% reduction in ammonia emissions that was achieved with slurry injection in the present study is within the range observed in earlier studies in Scandinavia (Hansen et al., 2003; Rodhe & Etana, 2005), but would probably have been larger if the slurry had actually been injected below the soil surface, rather than being applied manually after slots were created. Earlier studies evaluating different slurry injectors have found large variations in the degree of reduction in ammonia emissions, depending on soil conditions and function of the injectors (Hansen et al., 2003; Misselbrook et al., 2002; Nicholson et al., 2018; Rodhe & Etana, 2005). Rodhe and Etana (2005) reported a reduction in ammonia emissions of 52% relative to band application for the most effective type of slurry injector (making a 40–50 mm deep open slot), while the least effective injector showed no reduction. In a Danish study evaluating different slurry injectors (Hansen et al., 2003), ammonia emissions were found to be reduced by 20–75% compared with band application. Hansen et al. (2003) found that ammonia emissions reduction potential was linearly correlated to the volume of the slots created by the injectors, and concluded that high reduction potential is dependent on creation of slots with sufficient volume to contain all the slurry applied.

A potential trade-off with slurry injection that cannot be neglected is the possible increase in nitrous oxide emissions reported e.g. by Emmerling, Krein, and Junk (2020) and Duncan, Dell, Kleinman, and Beegle (2017). Other studies have reported no increase in nitrous oxide emissions (Fangueiro et al., 2017) or differing results (increase and no increase) between years (Seidel et al., 2017) and between crops (Fangueiro et al., 2018). Although emissions of nitrous oxide need to be minimised from a climate perspective, from an agronomic perspective the reported nitrous oxide emissions from slurry injection are small (less than 2% of total applied nitrogen) (Fangueiro et al., 2018; Duncan et al., 2017; Seidel et al., 2017) compared with the nitrogen saved in the form of reduced ammonia emissions.

#### 4.1.4. Effect of slurry acidification

The ammonia emissions abatement from slurry acidification was similar for all slurry types (75–88%), as could be expected since the reduction in pH was similar (Table 2). In other studies (Fangueiro et al., 2017, 2018; Seidel et al., 2017), acidification to pH 6.0 or below effectively reduced ammonia emissions. This was also the case in the present study, both for CS and LF acidified to pH 6.0 and for BD with a higher pH of 6.7. In a study by Wagner, Nyord, Vestergaard, Hafner, and Pacholski (2021), with pH reductions after field acidification comparable to those in the present study, the ammonia emissions reduction was smaller for anaerobic digestate, but similar for cattle slurry.

The higher initial emission rate from acidified BD (Fig. 2) compared with LF and CS can be explained by its higher pH

value. The very low emissions during the remainder of the experiment were probably caused by a combination of factors reducing the remaining amount of TAN available for emission, i.e. high initial emission rate depleting the slurry TAN pool and quick infiltration due to low slurry DM content.

For acidified CS, the pronounced increase in emission rate after around 48 h (Fig. 2) is most likely explained by an increase in slurry pH, since no corresponding increase was seen in the non-acidified treatments and there was no extreme temperature increase at that time (Fig. S2). Several factors possibly contributed to a late increase in slurry pH, including microbial oxidation of slurry volatile fatty acids (VFAs) to carbon dioxide and the buffering capacity of slurry and soil (Sommer et al., 2003). Pronounced increases in ammonia emissions from acidified slurry several days after application have only been reported previously by Pedersen et al. (2022), who found that emissions were low during the first 120 h after application of cattle slurry, and thereafter increased.

#### 4.2. Modelling ammonia emissions with ALFAM2

Although a wind tunnel environment cannot be expected to mimic open-air mass transfer perfectly, it has been found to provide similar ammonia emission measurements to open-air studies in some cases. The analysis in the original ALFAM work did not find a clear difference between wind tunnel and micrometeorological measurements by different research groups (Søgaard et al., 2002, Table 2). Sommer and Misselbrook (2016) reviewed cases involving active adjustment of wind tunnel air flow rate to match open-air wind speed close to the ground and found that this approach yielded similar emissions values to the Integrated Horizontal Flux (IHF) mass balance method. In the present study, the difference in cumulative emissions between wind tunnel measurements and ALFAM2 model predictions was small on average (see Section 3.2). High temperatures at the time of slurry application (Table 1) could have contributed to this better match, since recent experiments comparing wind tunnels with micrometeorological measurements have shown that under high-emission ambient conditions (in that case high temperature and wind speed), the two methods yield similar cumulative emissions (Hafner, Kamp, & Pedersen, 2023).

Differences between measured ammonia emissions and those predicted with the ALFAM2 model reflect several factors, including differences between wind tunnel measurements and the micrometeorological measurements used to estimate most model parameters, limitations in model structure, inaccurate parameter values, measurement error and effects not included in the model. With parameter values based on hundreds of plots from several countries, model evaluation with measurements from a single field study is not a sufficient basis for making changes to the model. However, the results in the present study indicate some processes that could benefit from additional attention.

Despite the importance of interactions between slurry and soil (Sommer et al., 2003, Sommer, Jensen, Clausen, & Søgaard, 2006), soil properties are not included as ALFAM2 predictor variables. This absence undoubtedly contributes to model error in general (Hafner et al., 2019) and possibly also in the present study. This limitation could be addressed by inclusion

of soil effects in a new parameter set, but the large field studies necessary for isolating soil effects have not been carried out to date.

The effect of anaerobic digestion of slurry on ammonia loss is an important topic because of a recent increase in the practice in some regions (Adamsen & Hafner, 2021; Statistics Sweden, 2020). According to model parameters (Hafner et al., 2021) and interpretation of emissions measurements (Chantigny et al., 2009; Evans et al., 2018; Neerackal et al., 2015), the reduction in DM and increase in pH due to digestion have opposing effects on ammonia emissions. In experiment 4, BD had much lower DM and much higher pH compared with CS but measurements showed little difference in cumulative emissions, implying that the two changes practically cancelled each other out. In model predictions the DM reduction effect dominated, leading to over-prediction of the difference between BD and CS. Improving the ability of the model to capture these important effects should be a goal of future work.

An increase in ammonia emissions flux over time in the absence of an increase in temperature or wind speed, as seen for acidified CS, is incompatible with the ALFAM2 model structure. If this phenomenon (see Section 4.1.4) is found to be widespread following acidification, model structure would need to be changed to enable more accurate predictions. Although slurry surface pH is known to change following application, incorporation of these dynamics into a simple model seems implausible (Hafner, Montes, & Rotz, 2013; Pedersen et al., 2022).

In experiments 1, 3 and 4, the ALFAM2 model predicted much higher initial flux than shown by measurements. The wind tunnel system can underestimate ammonia flux in the first measurement cycle in some cases (Pedersen et al., 2020), and therefore it is not possible to determine the magnitude of model error during these periods. The pattern with high initial flux was associated with later underestimation due to depletion of TAN from the “fast” pool in the model. Within the model, the pattern was caused by very high values for the emission rate constant from the “fast” pool ( $r_1$ ), caused in turn by somewhat higher air temperature and initial pH values (see Sections 2.1.1 and 2.1.2). Although parameter values were originally determined from measurements (Hafner et al., 2021), the comparison shown here suggests that the nature of the response ( $\log_{10}(r_1)$ ) is directly proportional to wind speed, temperature and pH) should be examined.

Some of the apparent error in model predictions is undoubtedly due to the substitution of wind tunnel average air velocity for open-air wind speed, or stated differently, problems in taking wind tunnel results as representative of open-air emission (Section 1.4). Despite some evidence that wind tunnels which match ambient air speed can replicate micrometeorological results (Sommer & Misselbrook, 2016), average velocity may differ from speed, especially with highly turbulent flow. A comparison of ethanol evaporation with the same wind tunnels as used in the present study showed that the average wind velocity within the tunnels corresponds to a much higher numerical value of air speed in open air in terms of mass transfer (Pedersen et al., 2020). This suggests that the model may underestimate wind tunnel emission rate, but the opposite was actually observed. Considering all this, careful

evaluation of the magnitude and nature of wind speed or wind tunnel airflow on emission rate is warranted.

The ALFAM2 model consistently underestimated the effect of acidification in reducing ammonia emissions in the wind tunnel experiments described here (Section 3.2). Unfortunately, emission reduction is not precisely related to pH or the change in pH (Nyord, Hafner, Adamsen, & Sommer, 2021) and resulting model parameters show high variability (Hafner et al., 2021).

## 5. Conclusions

As hypothesised, this study revealed an effect of slurry DM content on ammonia emissions, with a strong tendency for lower ammonia emissions (20% lower,  $p = 0.078$ ) from LF than from CS. For BD, the low DM content was counteracted by high pH, and thus cumulative ammonia emissions were not different from CS. The performance of the trailing shoes was affected by a hard, compact soil surface and the ammonia abatement compared with trailing hoses was non-significant for LF (16%) and BD (7%). For CS the ammonia abatement was greater (27%) and statistically significant, most likely related to the higher slurry DM content reducing the spreading out of slurry beside the trailing shoe soil slots. As hypothesised, slurry injection into 50 mm deep open slots reduced ammonia emissions compared with trailing hoses (by 37%) and slurry acidification effectively reduced ammonia emissions for all slurry types, by 75–88%. An unexpected increase in emission rate from acidified CS was seen from 48 to 70 h after slurry application, indicating an increase in slurry pH. The understanding of differences in ammonia flux patterns and cumulative emissions would benefit from further investigation of changes in slurry surface pH after field application, especially for acidified slurries. The effect of the structure of the uppermost soil layer (e.g. crust formation on dry clay soils) on slurry infiltration and ammonia emissions also needs further investigation.

Comparison of emissions measured in wind tunnels and values predicted by the ALFAM2 model showed reasonable agreement for cumulative emissions, but poorer model performance in predicting emission dynamics in some cases. Although this assessment of the model must be tempered by problems in relating wind tunnel measurements to open-air emissions, future work on the ALFAM2 model might benefit from an evaluation of air temperature, airflow and slurry pH effects.

## Funding

This work was funded by Cattle Foundation Skaraborg, Region Västra Götaland and the Swedish Board of Agriculture.

## Author contribution

Karin Andersson: Planning and preparation, Field experiments, ALFAM2 modelling, Data analysis, Writing – original draft (except Sections 3.2 and 4.2), Writing – review and

editing, Visualisation. Sofia Delin: Funding, Planning and preparation, Field experiments, Writing – review and editing. Johanna Pedersen: Planning and preparation, Data analysis (support), Writing – review and editing. Sasha Hafner: ALFAM2 modelling, Writing – original draft (Sections 3.2 and 4.2), Writing – review and editing, Visualisation. Tavs Nyord: Planning and preparation, Writing – review and editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

The authors would like to thank technicians Heidi Grønbaek and Per Wiborg Hansen at Aarhus University for their invaluable help with transporting the equipment to Sweden and setting up the first experiment. We would also like to thank the technical staff at Lanna research station, particularly Mattias Gustafsson and Erik Zakrisson, for practical help with the field experiments. Special thanks also to Johannes Forkman for valuable help with statistical questions.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.biosystemseng.2023.01.012>.

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