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Ammonia emission measurement with an online wind tunnel system for evaluation of manure application techniques

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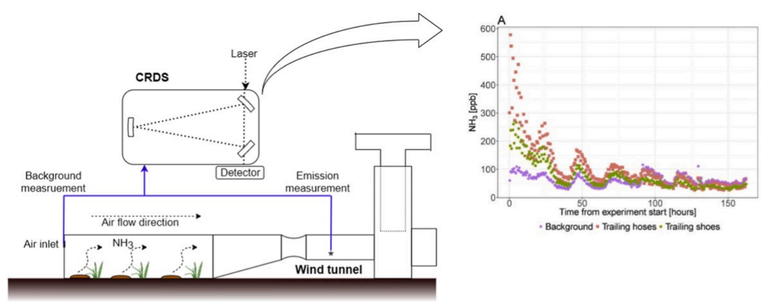
HIGHLIGHTS

- A system consisting of dynamic chambers and online measurement of ammonia was developed.
- The system allows for precise measurements with an average CV of 13% among triplicates.
- Ammonia emission abatement with trailing shoe was found to depend on soil type.
- Applying slurry at soil surface gave a large reduction in emissions.

GRAPHICAL ABSTRACT



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ABSTRACT

Field application of liquid manure contributes substantially to atmospheric ammonia. Low emission application methods are commonly used to reduce ammonia transfer to the atmosphere. To document which application method results in lower ammonia volatilization there is a need for high precision measurements to ensure that small differences in total emission and emission patterns can be quantified. This paper presents the evaluation and application of a new system of dynamic chambers (wind tunnels) with online cavity ring down spectroscopy measurements of ammonia. The system allows for high time resolution of 104 min throughout the measuring period ($\geq$90 h) when testing two treatments and one reference in triplicates. Measurement variability is low with a coefficient of variation of 13 $\pm$ 8% within triplicates. The system was used to investigate the effect of trailing shoes compared to trailing hoses on different soil and crop types, where the expected differences in ammonia volatilization are low. The results show that when applying pig slurry on coarse sand a significant reduction of 47 $\pm$ 20% was obtained, whereas the reduction when applied on loamy sand and sandy loam was lower and occasionally insignificant. During ten experiments on three different soil types, an overall average reduction of ammonia volatilization from using trailing shoes compared to trailing hoses was found to be 19 $\pm$ 12%. Furthermore, the importance of correct use of trailing hoses was examined by comparing with application above the crop canopy. Application at the ground surface gave an ammonia emission reduction of 40 $\pm$ 13% compared to application 20 cm above the canopy.

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1. Introduction

Intensive livestock production yields vast quantities of manure, a valuable by-product if utilized correctly due to the content of important nutrients. Good manure handling practices are important to ensure proper use of the nutrients and reduce the risk of polluting the surroundings. Emissions from manure include greenhouse gases, ammonia, and odor. The main sources of emissions are livestock production units, storage facilities, and land application. These contribute to environmental pollution due to nitrogen deposition, acidification, global warming (Eurostat, 2017; Haisler and Jacobsen, 2017), and formation of particles (Walker et al., 2006), which are associated with negative health impacts (Eurostat, 2017). The emissions depend on several factors such as meteorological conditions, soil and slurry conditions, crop type, application method, timing of application, and the interactions of these parameters.

Agriculture is the main source of ammonia emissions in the EU-28 countries (Eurostat, 2017) and accounts for 75% of the global NH3 emissions (Van Vuuren et al., 2011). Nitrification of ammonia is one of the most important contributors to acidification of the environment, and accounted for the highest share of acidifying potential in the EU-28 countries in 2014 (Eurostat, 2017). In addition, chronic deposition of nitrogen is linked to reduction of biodiversity in e.g. grasslands (Stevens et al., 2010). Mitigating ammonia emissions is a key strategy for preventing environmental acidification. The EU-28 countries have committed to reduce emission of ammonia from agriculture in order to reduce the environmental impact of food production (Haisler and Jacobsen, 2017). Reducing emissions from field application is an essential step towards overall ammonia reduction, since field application contributes 42% (Eurostat, 2017) of total agricultural ammonia emissions in the EU.

2. Ammonia emissions

Extensive research has been done on ammonia emissions from field-applied slurry. Different measuring methods have been used, of which the most common are micrometeorological methods (Häni et al., 2016; Misselbrook et al., 2005a; Pacholski et al., 2006; Mannheim et al., 1995); enclosure methods using various designs of static and low-flow dynamic chambers (Parker et al., 2013); and wind tunnels, arguably a type of dynamic chamber, but with high and primarily longitudinal air flow (Bell et al., 2015; Lockyer, 1984; Mannheim et al., 1995; Rochette et al., 2008; Sommer and Misselbrook, 2016).

Wind tunnels require smaller plots than micrometeorological methods, which makes it possible to have more replicates. Because they modify the measurement environment and have been observed to overestimate ammonia emissions compared to micrometeorological mass balance measurement techniques (Misselbrook et al., 2005b; Sommer and Misselbrook, 2016), they may not be suitable for determining absolute emission under natural conditions. Nonetheless, the small area footprint required and the possibility to make unbiased replications make the wind tunnel method an appealing option for comparative measurements if designed correctly (Misselbrook et al., 2005a, 2005b; Sommer and Misselbrook, 2016). With wind tunnels, ammonia is typically captured by bubbling exhaust air through acid impingers and later quantified (Bell et al., 2015; Lockyer, 1984; Rochette et al., 2008; Smith and Watts, 1994). This method is laborious, as the acid impingers have to be changed manually and acid solutions saved and later analyzed in a laboratory, leading to limited time resolution (median $>$ 11 h (ALFAM2)) and little information about the ammonia emission dynamics. The design of the wind tunnels strongly affects results, with air flow or air velocity being recognized as the most important factor in several studies, as higher values result in higher measured fluxes (Eklund, 1992; Smith and Watts, 1994; Sommer and Misselbrook, 2016). Eklund (1992) argued that the optimal air velocity depends on the tunnel design and the source of emissions. The air flow should be sufficiently high to provide realistic levels of turbulence above the emitting surface. Despite this knowledge, limited effort has been brought into investigating air flow. Two different approaches have been used: setting the air flow to a constant value (Bell et al., 2015; Bhandral et al., 2009; Smith et al., 2000) or adjusting the air flow so average air velocity matches ambient wind speed during the experiment or short measurement intervals (Braschkat et al., 1997; Mannheim et al., 1995). In wind tunnel studies air flow is commonly reported as average air velocity in the longitudinal dimension [m s^{-1}]. This value is either calculated from the volumetric flow rate and cross-sectional area (i.e., the average air velocity) or measured at one point in the emission chamber using an anemometer. Considerable variations in air velocity throughout the tunnel chamber will undoubtedly occur (Jiang et al., 1995). A single report of air flow does not provide any knowledge of the velocity profile and potential variations throughout the chamber or at the soil surface where the emissions occur.

Several factors influence ammonia emission from field applied slurry including total ammoniacal nitrogen (TAN) concentration (Huijsmans et al., 2018), incorporation into soil (Hafner et al., 2019; Rodhe et al., 2004; Smith et al., 2000), application technique (Hafner et al., 2019; Rodhe et al., 2004; Smith et al., 2000), application rate (Hafner et al., 2019; Huijsmans et al., 2018), slurry pH (Sommer and Olesen, 1991), slurry dry matter (Hafner et al., 2019; Huijsmans et al., 2018; Sommer et al., 2006), air temperature (Bell et al., 2015; Hafner et al., 2019; Huijsmans et al., 2018), wind speed (Hafner et al., 2019; Huijsmans et al., 2018; Misselbrook et al., 2005b), rainfall rate and timing (Hafner et al., 2019; Martínez-Lagos et al., 2013), crop conditions (Huijsmans et al., 2018; Smith et al., 2000), and soil type and conditions (Bell et al., 2015; Huijsmans et al., 2018; Smith et al., 2000). The soil conditions influence infiltration together with manure dry matter and application technique. Infiltration is often considered to be highly important for emissions (de Jonge et al., 2004; Hafner et al., 2019; Misselbrook et al., 2005b; Rochette et al., 2008).

Although comprehensive research has been conducted on ammonia emissions from field-applied slurry, a number of questions remain concerning the factors that influence emission and the relative importance of these. The primary aim of this study was to develop and evaluate a new wind tunnel system with online measurements. Online measurements allow for higher time resolution and insight into temporal ammonia emission dynamics. As a part of the wind tunnel evaluation, the air-side mass transfer velocities (Lee et al., 2004; Schwarzenbach et al., 2003) in the wind tunnel system have been compared to outside conditions via measuring the evaporation of a pure liquid (ethanol). The air-side transfer velocities are assumed to depend only on the turbulence intensity (at the same temperature) and therefore reflect whether turbulence intensities in the wind tunnels are comparable to natural outside conditions. A similar approach was used by Parker et al. (2013). A secondary aim was to use the new system to examine the interaction between soil type and ammonia volatilization from slurry application with trailing shoes and trailing hoses. The objectives were to: (i) Optimize a wind tunnel system measuring ammonia with continuous online measuring technique, (ii) Develop a method for evaluation of the air exchange rate (turbulence intensity) in the emission chamber, (iii) Conduct comprehensive tests on the effect of trailing shoes and trailing hoses on ammonia emissions by including soil type as a factor, and (iv) Illustrate the importance of correct use of slurry application methods by measuring ammonia emissions from slurry applied by trailing hoses at the grass canopy and from 20 cm above the grass canopy.

3. Materials and methods

3.1. Measuring system

3.1.1. Wind tunnels

Emissions were measured using nine wind tunnels operated as dynamic chambers with a continuous and constant air flow (Fig. 1). The

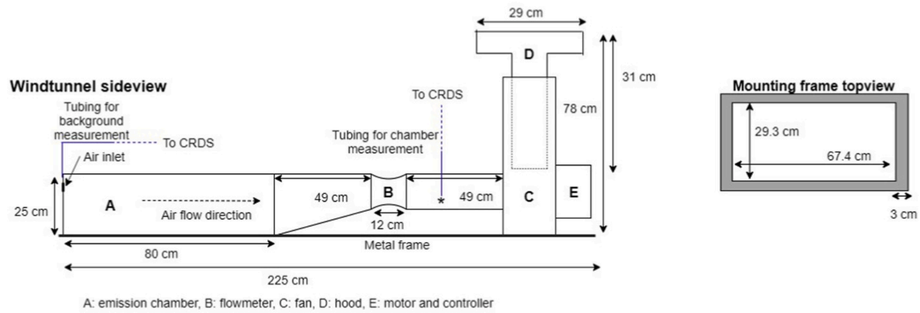

Fig. 1. Sketch of wind tunnels, not to scale.

wind tunnels consisted of a rectangular open-bottomed stainless steel chamber (80 × 40 × 25 cm). The chamber was connected to a fan (SEAT 20 (tunnel 1) and SEAT 25 (tunnel 2–9), SEAT ventilation, Verniolle, France) via a steel duct. An orifice flow meter (FMU 80-63, Lindab, Haderslev, Denmark), where the volumetric flow is calculated from measured pressure drop, was included in the duct. Each fan was run by an electric motor (MS 71B-B34, BUSCK, Kållered, Sweden) and a frequency converter (ATV12H037M2, Schneider Electric, Rueil-Malmaison, France). A micro manometer (5825, DP-CALCTM, Shoreview, MN, USA) was used to measure the air flow through the tunnel, which was manually adjusted with the frequency converter to an air exchange rate of 25 min^{-1}. The air exchange rate corresponds to a calculated mean air velocity of 0.33 m s^{-1} in the emission chamber. Resistance of the air flow through the chamber and stabilization of the fans was increased by having a small air-inlet (33.5 × 1.3 cm) into the tunnels and a hood at the air exit of the fan. The small inlet prevents back-flow which might lead to false emission measurements due to erroneously high background concentrations. To control the amount of slurry for each tunnel and avoid leaks, a seal between the soil and tunnel was obtained by a metal frame inserted 40 mm into the soil for the tunnel to be mounted on. The frame gave a plot area of 0.2 m^2.

Air from the tunnels was drawn through 8 mm PTFE tubing with a minimum flow rate of 0.9 L min^{-1} to a channel selection manifold consisting of 19 on/off valves (P/N 038T2S24-54-4, Bio-Chem Fluidics, Boonton, NJ, USA) with a valve for each sampling tube. The tubing was heated to approximately 40 °C by heating cables and insulation pipes. The valve manifold was controlled by a custom-built data logger. Air was drawn from each valve for 8 min. A cavity ring down spectroscopy (CRDS) instrument (G2103 NH3 Concentration Analyzer, Picarro, CA, USA) was connected to the valve manifold for continuously measuring ammonia.

3.1.2. Instrumentation

The recovery of ammonia throughout the system was tested in the field with a standard ammonia gas (11.3 ppm, Linde, Surrey, UK) added to the tube inlets. The recovery of ammonia within the 8 min of measurement interval measured at several occasions out in the field was minimum 90%. In experiments A and B, a leakage in the connection to the CRDS was discovered at the completion of experiment B. The data in experiments A and B has therefore been corrected for the decreased recovery measured onsite with the reference gas.

3.1.3. Selection of air flow rate

To assess the air-side mass transfer velocity (mass transfer coefficient) and hence turbulence intensity, evaporation rates of ethanol inside the tunnels were compared to outside evaporation at the same time. Nine petri dishes (78 mm diameter) were placed evenly in a three x three grid on the soil surface within the tunnel frame. 20 mL of ethanol (≥96% (v/v)) were added to the petri dishes. The tunnels were placed on top of the frames and run with a fixed air exchange rate for one or two h. Three tunnels were used for each air exchange rate. Simultaneously, six to nine petri dishes were placed next to the tunnels to measure evaporation outside. Evaporations with six different air exchange rates, from 15 to 47 min^{-1}, were measured (data not included). Based on these, an air exchange rate of 25 min^{-1} was chosen for all the experiments (see section 3.1.1. for discussion of this). Thereafter several experiments were performed with different ambient temperature and wind speed conditions on three tunnels with an air exchange rate of 25 min^{-1}. During these experiments, the average temperatures ranged from 6 to 17 °C and the average wind speed ranged from 2.7 to 7.5 m s^{-1} (Fig. S1).

3.2. Experimental setup and site

3.2.1. Soil

Field trials were performed in spring and summer 2018 at different sites at Research Center Foulum (Aarhus University, Tjele, Denmark). Experiments with winter wheat crops and bare soil were performed in an experimental field facility established in 1993 (Nyord et al., 2010). The facility has three soil types: coarse sand (Ortic Haplohumod), loamy sand (Typic Hapledult), and sandy loam (Typic Agrudalf) with clay contents of 4, 9, and 18% respectively (Chen et al., 2013). Experiments on clover grassland were performed on two different fields at Research Center Foulum, both with the same loamy sand soil as in the experimental field facility (the soil was established in 2015). Soil-water contents and dry bulk densities were determined gravimetrically using 100 cm^3 soil cores taken at 0–5 cm depth and 1:1 water pH was determined using the standard method (USDA, 2009). Each experiment required a soil plot of approximately 16 × 2.5 m. The tunnels were placed adjacent to each other, and differences between blocks of the same soil type were expected to be small. All soil information and analysis can be found in Table 1.

3.2.2. Slurry

Cattle and pig slurry was sampled from concrete slurry storage tanks at Aarhus University Foulum.

Analyses were performed using standard methods for dry matter content (APHA, 1999), total nitrogen (AOAC, 1999), and TAN (IS, 1984). All analysis results and application rates during the experiments can be found in Table 2.

3.2.3. Slurry application

To investigate application by trailing shoes a metal frame on wheels was constructed (Fig. 2a). Three trailing shoes (Bomech B. V., Albergen, The Netherlands) were mounted to the frame, with a distance of 25 cm between them. The force on each shoe was adjusted to the soil, with a maximum force at the end of each trailing shoe of approximately 117 N based on the maximum possible force typically applied on a commercial slurry tanker boom. The slits were obtained at a constant speed of approximately 2 km h^{-1}. The resulting slits from running the metal frame with the trailing shoes differed greatly in size and geometry due to the different pressure applied, soil-water content, and crops.

At the start of the experiments a pre-determined volume of the slurry

Table 1
Soil properties for all the experiments and force on each trailing shoe when applicable. Standard deviations are displayed in parenthesis ($n = 3$).

Exp	Soil	Crops	Dry bulk density [g cm^{-3}]	Water content [g g^{-1}]	pH	Force on trailing shoe [N]
A	Coarse sand	Winter wheat	1.41 (0.08)	0.09 (0.004)	4.5	78
B	Loamy sand	Winter wheat	1.32 (0.07)	0.13 (0.01)	4.4	88
C	Sandy loam	Winter wheat	1.41 (0.06)	0.17 (0.01)	6.3	98
D	Loamy sand	Grass	1.15 (0.08)	0.15 (0.01)	4.9	117
E	Loamy sand	Grass	1.15 (0.08)	0.21 (0.01)	4.8	117
F	Coarse sand		1.41 (0.08)	0.11 (0.004)	4.5	88
F	Loamy sand		1.32 (0.07)	0.18 (0.01)	4.9	108
F	Sandy loam		1.41 (0.06)	0.18 (0.01)	6.4	117
G	Coarse sand		1.41 (0.08)	0.08 (0.002)	4.6	
G	Loamy sand		1.32 (0.07)	0.13 (0.01)	5.5	
G	Sandy loam		1.41 (0.06)	0.14 (0.005)	6.8	
H	Loamy sand	Grass	1.61 (0.08)	0.15 (0.01)	5.9	117
I	Loamy sand	Grass	1.61 (0.08)	0.19 (0.01)	6.1	
J	Loamy sand	Grass	1.61 (0.08)	0.20 (0.02)	6.2	

Table 2
Slurry properties for all the experiments. Standard deviations are displayed in parenthesis ($n = 2$).

Exp	Type	Application rate		Dry matter [%]	Total N [g L^{-1}]	Ammoniacal N [g L^{-1}]	pH
		[kg m^2]	[g NH_4-N m^{-2}]				
A	Pig	4.5	10.00 (0.14)	3.87 (0.05)	2.62 (0.42)	2.22 (0.03)	7.19 (0.04)
B	Pig	4.5	9.90 (0.38)	3.84 (0.09)	3.45 (0.23)	2.20 (0.08)	7.04 (0.03)
C	Pig	4.5	10.11 (0.44)	3.40 (0.18)	2.92 (1.07)	2.25 (0.10)	7.10 (0.08)
D	Cattle	3.5	9.77 (0.44)	9.04 (0.01)	5.07 (0.17)	2.79 (0.13)	6.94 (0.01)
E	Cattle	3.5	9.90 (0.68)	8.96 (0.02)	4.61 (1.04)	2.83 (0.19)	6.94 (0.01)
F	Pig	4.5	5.21 (0.55)	3.17 (0.27)	1.71 (0.10)	1.16 (0.12)	7.30 (0.01)
G	Pig	4.5	5.64 (0.08)	3.02 (0.01)	1.85 (0.04)	1.25 (0.02)	7.64 (0.01)
H	Cattle	3.5	9.51 (0.05)	8.63 (0.02)	4.26 (0.27)	2.72 (0.01)	6.83 (0.01)
I	Cattle	4.5	15.77 (0.06)	6.76 (0.01)	3.05 (0.04)	3.50 (0.14)	7.37 (0.04)
J	Cattle	4.5	15.93 (0.87)	6.78 (0.07)	3.02 (0.16)	3.54 (0.19)	7.33 (0.04)

(Table 2) was applied manually by a hose attached to a watering can (as done by (Bell et al., 2015; Misselbrook et al., 2005b; Wulf et al., 2002)) evenly in the three slits when mimicking application by trailing shoes, at the soil surface when mimicking application by trailing hoses, or from 20 cm above the grass canopy when mimicking application by trailing hoses used with a distance between the trailing hoses and the ground.

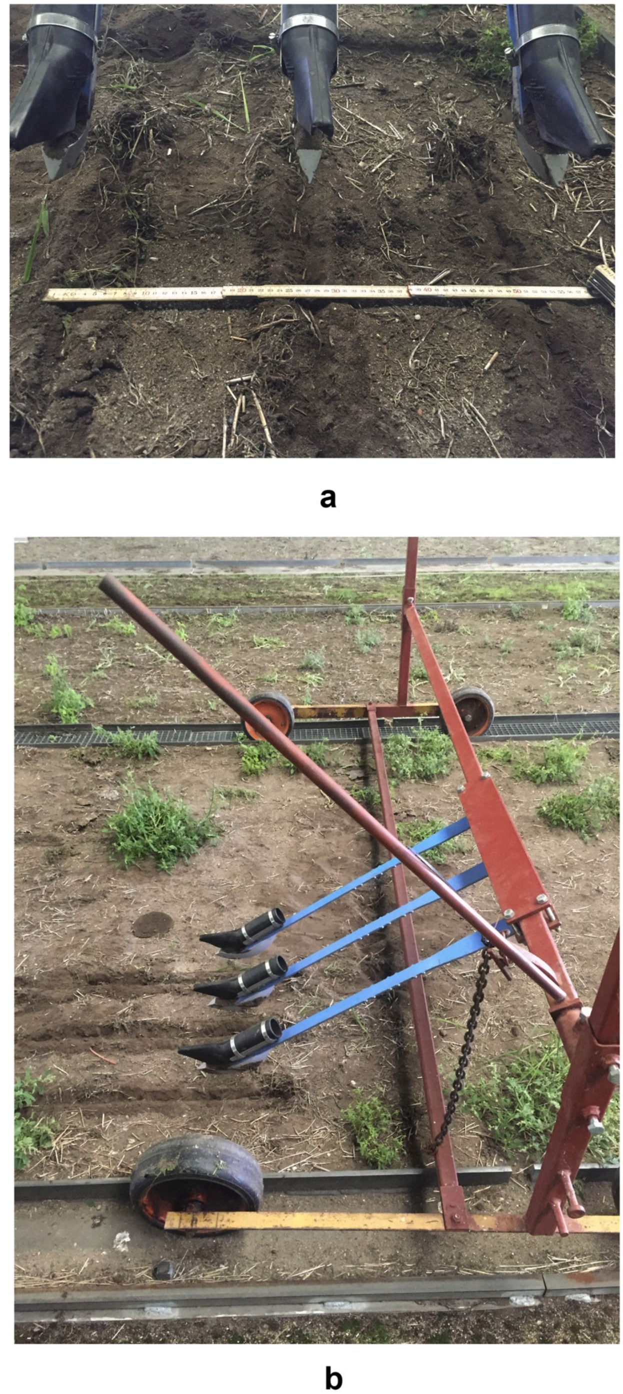

Fig. 2. (a) Metal frame with Bomech trailing shoes attached. (b) Slits made by Bomech trailing shoes on coarse sandy soil.

The slurry was thoroughly stirred before the amount needed was removed and applied.

3.2.4. *Meteorological data*

A weather station (Theis CLIMA, Göttingen, Germany with a Campbell CR10xB data logger, Campbell Scientific, INC, UT, USA) was continuously measuring ambient air temperature (Hygro-Thermo Transmitter-compact, THEIS CLIMA) and soil temperature (Temperature Transmitter, Theis CLIMA) 5 cm from the soil surface in 10 min intervals. Average weather data can be found in Table 3.

Table 3
Soil and air temperature during all experiments. Averages of the first 6 and 24 h after application and total experimental period. Soil temperatures are measured at 5 cm depth.

	Air temperature [°C]			Soil temperature [°C]		
Exp	6 h avg.	24 h avg.	Total avg.	6 h avg.	24 h avg.	Total avg.
A	25.1	18.9	15.1	20.4	17.6	15.9
B	22.6	17.6	18.2	19.8	16.8	16.7
C	21.9	20.2	20.4	17.3	16.8	16.8
D[a]	15.7	12.8	15	NA	16.3	17.1
E	22.5	17	16.9	24.1	18.6	18.3
F[b]	21.6	19.3	17.6	19.2	18.5	17.5
G[b]	23.5	19.6	15.6	18.5	17.7	15.7
H	15.9	14.7	15.7	19.4	17.5	17.7
I	10.4	9.2	10.4	10.7	9.1	10.6
J	15	12.9	15	12.1	11.1	12.1

[a] Soil temperature is missing for the first 10 h.
[b] Soil temperature measured in loamy sand.

3.3. Data treatment

The experiments varied in duration from 90 to 157 h. To compare the accumulated emissions they were calculated for 90 h. For all of the experiments, most of the emissions had occurred at this time (minimum 83% of total emissions during the measuring period, data not included). For the experiments with data for >90 h the emissions were at such a low level that no differences in ammonia emissions from the different treatments were observed (data and analysis not included).

The volatilization flux of ammonia in units of g m^{-2} min^{-1} was calculated from the concentration, the air flow in the tunnel, and the area of the soil surface covered by the tunnel (Equations (1) and (2)).

$$F_{NH3} = (C^*q) / A \quad (1)$$

where F_{NH3} is the flux (g min^{-1} m^{-2}), C is the concentration (g L^{-1}), q is the volumetric air flow rate (2016 L min^{-1}), and A is the area of the soil surface within the tunnel (0.2 m^2).

Concentration was converted to units of g L^{-1} and corrected for background concentration by Equation (2):

$$C = P / (R^*T)/(c_o - c_i) ^*M \quad (2)$$

where C is the ammonia concentration (g L^{-1}) (average of the last 30 s of a measuring cycle), P is the pressure (1 atm), R is the gas constant (0.08206 L atm K^{-1} mol^{-1}), T is the temperature (K), c_o is the outlet concentration (ppb), c_i is the inlet (background) concentration (ppb), and M is the molar mass of ammonia (17.03 g mol^{-1}).

To calculate emission the flux between two measurements was taken as the value calculated by equation (1) at the beginning of the 104 min interval (i.e., a left Riemann sum). When concentrations are not significantly higher than the background, they are set to 0.

During five of the experiments, an error occurred in the sampling system, resulting in loss of emission data. The missing data spans from 4 h (E) to 16.6 h (F) (Fig. S3). The missing data were estimated to obtain more correct cumulative emission calculations. Linear interpolation was used to estimate the data if the ambient air temperature was either increasing or decreasing during the period. If the temperature was increasing followed by a decreasing or vice versa, a minimum or maximum was estimated based on the time of day, the temperature and the temperature-emission ratio in the dataset. Then linear interpolation was used from the points right before and after the missing data and the estimated maximum or minimum. The standard deviations of the missing data were calculated from the highest coefficient of variation in the data before and after the missing data.

3.3.1. Statistics

Single factor or two factor analysis of variance (ANOVA) was used to test for differences among application methods. Within each block, one tunnel with each treatment was present and randomly assigned. Data were analyzed with an ANOVA, with the cumulative emission after 90 h as the response variable and an individual plot in a single wind tunnel as an observational unit. Subsequently, Tukey's test, with a confidence interval of 95%, was used to investigate which treatments were significantly different. Only a single soil type was used in Experiments A, B, and C (Table 1). Data from these three experiments were analyzed together in a two-way factorial ANOVA with soil type and application method as the independent variables, based on the assumption that differences (including any interactions) among experiments were primarily related to soil type. Experiments F and G were analyzed separately with one-way ANOVA with soil type as the independent variable. Experiments D and E were analyzed together with a one-way ANOVA with application method as the independent variable and experiment as a blocking variable. Due to a large difference in emissions from experiment H compared to I and J, the results from H were analyzed separately with a one-way ANOVA with application method as the independent variable. Experiments I and J were analyzed together with a one-way ANOVA with application method as the independent variable and experiment as a blocking variable.

A paired *t*-test was used to test the average reduction of ammonia emissions using trailing shoes compared to trailing hoses in all the experiments.

Graphics include error bars representing one standard deviation.

4. Results and discussion

4.1. Evaluation of wind tunnels

4.1.1. Air exchange rate

Theoretically, ammonia emissions from an aqueous surface are expected to be determined almost completely by air-side resistance due to its relatively low partitioning into the gas phase (Hafner et al., 2012; Schwarzenbach et al., 2003) partly as a result of the rapid equilibrium between NH_3 and NH_4^+. Consequently, the emission is expected to depend on turbulence intensity, which in turn is related to air velocity. Observations indeed support that emissions are highly depending on air velocity (Huijsmans et al., 2018; Misselbrook et al., 2005c; Sommer and Misselbrook, 2016), and wind tunnel air exchange rate is therefore an important operating parameter. The optimal air exchange rate inside the tunnel should create turbulence at the soil surface comparable to what is found outside the tunnels, and should be relatively homogeneous throughout the tunnel. Parker et al. (2013) presented a methodology based on evaporation of water for standardizing and comparing different evaporation chambers and correlate the emissions within the chamber to field conditions. Inspired by this method, evaporation measurements of ethanol inside and outside the emissions chambers were used as a method for rapid and quantitative evaluation of the sweep air flow. Ethanol was chosen rather than water vapor, since it evaporates more quickly and has a low atmospheric concentration (compared to water).

Based on testing several different air exchange rates, 25 min^{-1} was chosen as the optimal. It gave rather homogeneous evaporation throughout the soil surface in the emission chamber, and the evaporations were close to the evaporations outside the tunnels. Evaporation tests at different days allowed to test under several different weather conditions. Under these very different weather conditions, the emissions inside and outside the tunnels where in the same range (Fig. 3) with an average difference of 11 ± 7%. The air exchange rate of 25 min^{-1} corresponds to a calculated mean air velocity of 0.33 m s^{-1} in the emission chamber. Jiang et al. (1995) did extensive testing of velocity profiles with an anemometer within a wind tunnel emission chamber of the same dimensions as the ones used in this paper. They found that a velocity of 0.33 m s^{-1} gave the most stable velocity profile throughout the chamber.

It was assumed that the air flow through the tunnel was constant during the experiments. The air flows were measured and adjusted right

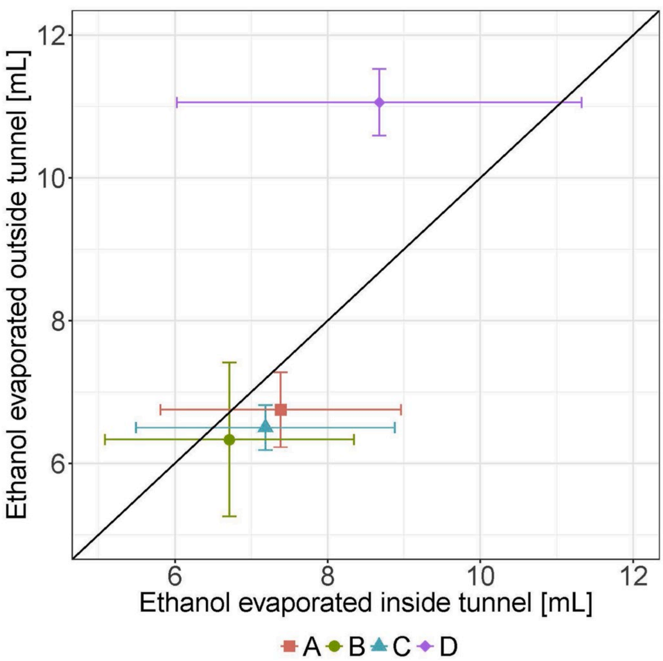

Fig. 3. Average evaporation of nine petri dishes inside a wind tunnel emission chamber ($n = 3$) compared to average emission from six petri dishes outside of wind tunnel. Standard deviations are displayed as error bars ($n = 27$ for inside measurements, $n = 6$ for outside measurements). The letters indicate test days with different weather conditions.

after slurry application and measured at the end of the experiment. Slight changes occurred, assigned to changes in the ambient wind speed. On two occasions, the difference compared to the air flow at the time of application was 7%, but in all other cases, the variations were within 5%.

It is concluded that the wind tunnel design and air flow rate used in this study provides a good simulation of outside turbulence intensities.

4.1.2. Sampling system

Online CRDS measurement provides data with a 104 min time resolution with one control and two treatments in triplicates (nine tunnels in total). This time resolution is very high throughout the measuring period compared to experiments with manually sampling (Bell et al., 2015; Rochette et al., 2008; Sommer et al., 2006). The conditions inside the dynamic chamber are in principle different from real field conditions, therefore the absolute emission values cannot necessarily be translated to outside emissions. As the effect of air velocity (turbulence at the soil surface) compared to ambient conditions are not very different, it might be possible to correct the chamber measurements in the future to yield absolute emission data. This has not been investigated further in this paper. Low variation in the results (Section 3.2) indicates that the method gives reliable quantitative data, which can be used to compare different application methods, slurry types or soil types. 23 sets of measured triplicates give an average coefficient of variation of 13 ± 8%. The variation is low compared to the literature (Nyord et al., 2012; Rochette et al., 2013), which is necessary when examining methods with expected effects in a lower range, such as trailing shoes compared to trailing hose. The system allows for experiments under realistic agricultural conditions with continuous measurements over long periods providing comprehensive datasets with details of the emission dynamics, such as diurnal patterns (Fig. 4).

The sampling system typically requires 5–8 min to reach a stable reading of both high and low concentrations. The response time depends on the internal surface temperature and ammonia concentration differences. In the first cycle of ammonia measurements, the concentration in most cases increased close to linearly due to the rapidly increasing emissions shortly after slurry application (Fig. S2). It cannot, however, be completely ruled out that the sampling system (tunnels, tubes, fitting, valve-block, etc.) needed more time to equilibrate in the first measurement cycle, which would have resulted in a minor underestimation of the first data point. From the second measuring cycle, all tunnels reached stable readings with a sampling time of 8 min (Fig. S2).

High emission measurements result in false high background

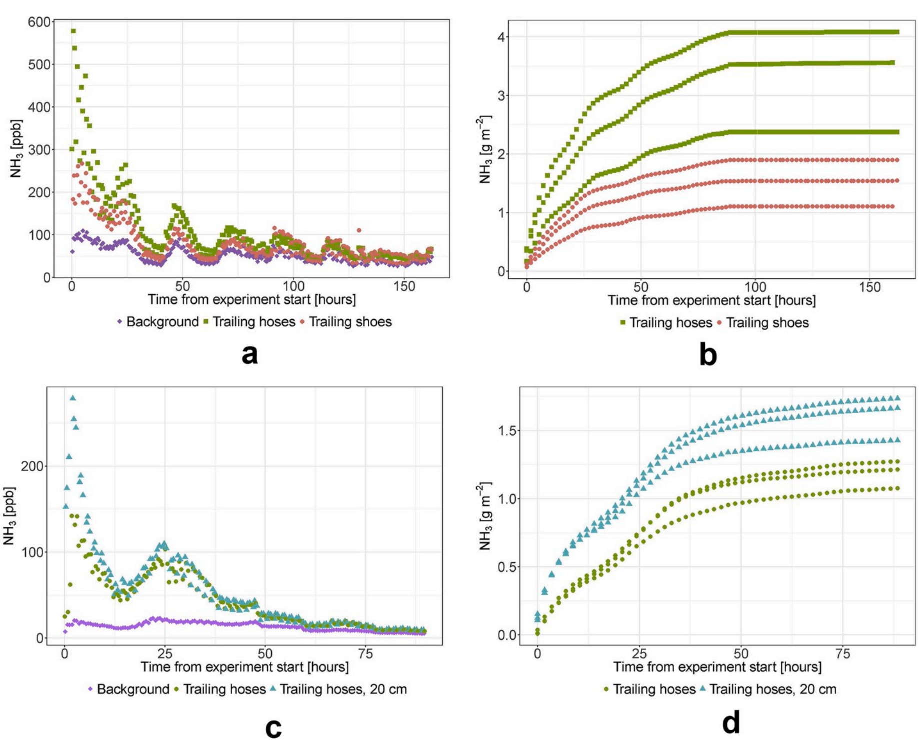

Fig. 4. Outlet ammonia concentration and accumulated loss of nitrogen due to ammonia volatilization during experiment A and I.

measurements as the system did not have time to reach a stable background reading. With emission measurements of 700 ppb before a background measurement, the underestimation of the ammonia concentration is approximately 3.5%, whereas an emission measurement of 300 ppb before a background measurement will lead to an underestimation of 1%. Generally, only very few measurements are above 300 ppb (Fig. 4), and it only occurred during the first couple of cycles. Therefore, the slightly higher background readings have a very limited effect on the overall results.

4.2. *Ammonia emissions from surface applied slurry in growing crops*

Ten experiments were performed testing the effect of trailing shoes compared to trailing hoses. In three experiments (A, B, and C) the two application methods were tested against each other on three soil types with winter wheat crops (Figs. 5a and 4). Two experiments (F and G) were conducted in which each application method was tested across the three soil types simultaneously (Fig. 5b). Trailing hoses and trailing shoes were compared on a clover grass field in two identical experiments (D and E) (Fig. 6a) and the last three experiments focused on comparing trailing hoses used correctly and trailing hoses applying slurry 20 cm above the grass canopy (H, I, and J) (Figs. 6b and 4).

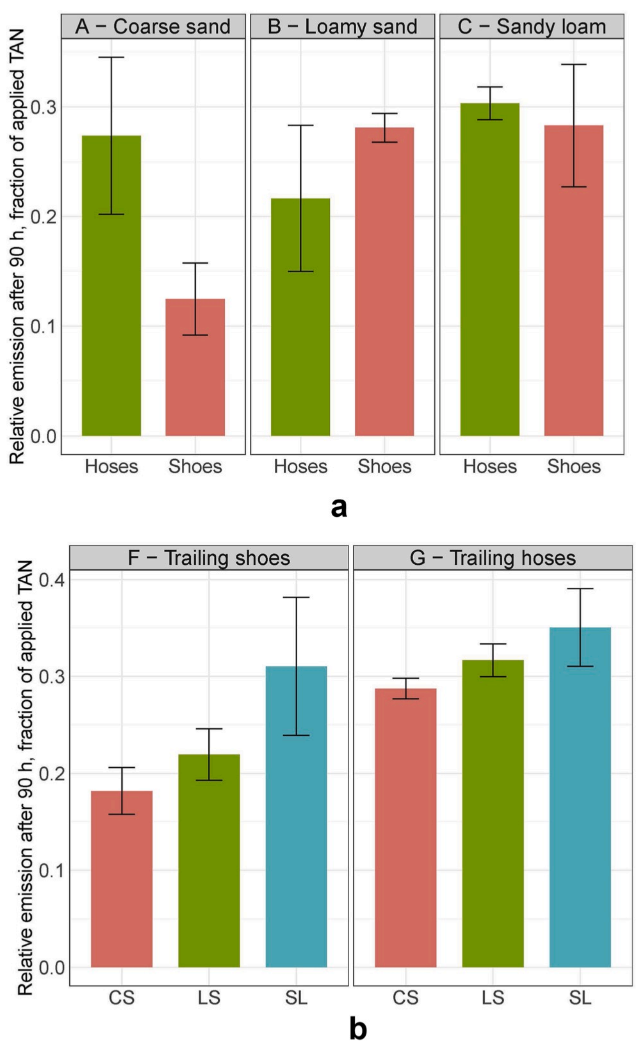

Fig. 5. Effect of slurry application method for pig slurry applied to coarse sand, loamy sand and sandy loam soils. Standard deviations are displayed as error bars ($n = 3$ for emission measurements and $n = 2$ for NH4-N analysis). (a) Three experiments varying soil type with trailing shoes (Shoes) and trailing hoses (Hoses). (b) Two experiments varying application method on three different soil types: coarse sand (CS), loamy sand (LS) and sandy loam (SL).

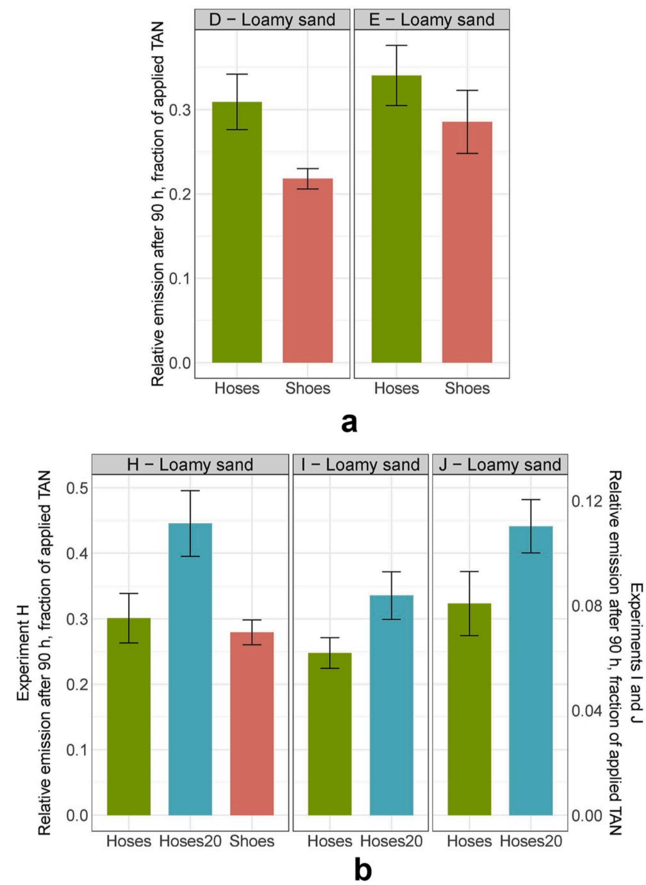

Fig. 6. Effect of slurry application method for cattle slurry applied to loamy sand soil with clover grass. Standard deviations are displayed as error bars ($n = 3$ for emission measurements and $n = 2$ for NH4-N analysis). (a) Two experiments with trailing shoes (Shoes) and trailing hoses (Hoses). (b) Three experiments with trailing hoses and trailing hoses lifted 20 cm (Hoses20) above the grass canopy. H includes trailing shoes.

Comparing trailing hoses to trailing shoes on the same soil with winter wheat (A, B, and C) a significant difference in the ammonia volatilizations was only found on coarse sand with a reduction of 56 ± 20% (A) ($p < 0.05$) (Fig. 5). Testing trailing shoes across three different soil types (F), significantly higher ammonia volatilization was found on sandy loam compared to loamy- and coarse sand ($p < 0.05$) (Fig. 5). The pattern of ammonia volatilization from trailing hoses across three soil types (G) was the same as with trailing shoes (F), but the soil type effect was markedly lower (Fig. 5). When trailing shoes are used on bare soils or soil with cereal crops it is used to create a slit for the slurry to be contained in. The intent is to create a lower surface area between the slurry and air, hence lowering the emissions compared to trailing hoses. On soils with high clay content, a higher mechanical force is needed in order to create the slit. Under certain conditions, slits might not be obtained as the force applied on each shoe is limited by the construction and weight of the boom. This limitation can explain why the abatement obtained on coarse sand is more efficient with an average reduction of 47 ± 20% compared to the loamy sand and sandy loam (average reductions of 2 ± 10 and 9 ± 3% respectively) (A, B, C, F, and G). Hence, the reduction of ammonia volatilization from slurry applied by trailing shoes is highly dependent on the soil type.

Air temperature has been observed to be one of the most important factors for ammonia emissions (Bell et al., 2015; Huijsmans et al., 2018; Martínez-Lagos et al., 2013). Relatively large variations in air temperatures occurred for experiments A, B, and C with an average air temperature of 15.1 °C and 20.4 °C during A and C, respectively. The high average air temperature during C was due to short periods with hot temperatures (above 27 °C). The average soil temperatures, as well as the 6 and 24 h averages of both air and soil temperature, were very similar, (Table 3). It is assumed that the experiments can be compared as most of the emissions occur during the first 24 h after application.

When applying slurry on grass fields with trailing shoes no slits are created due to the grass. When the trailing shoes move across the grass the force of the shoes ensures that the slurry is applied on top or a little below the surface of the grass. This presumably results in a lower surface exposed area of slurry and creates better conditions for slurry infiltration compared to trailing hoses. Lower ammonia volatilization was found in three experiments testing trailing shoes against trailing hoses on loamy sand with clover grass with an average reduction of 17 ± 4% (D, E, and H) (Fig. 6). The reduction is significant in two of the three experiments (D and E) ($p < 0.05$), but low compared to the reduction found on sandy soil with winter wheat (Figs. 5 and 6). The lower ammonia volatilization from trailing shoes compared to trailing hoses on grass is in contrast with findings by Smith et al. (2000). They found that the reduction of ammonia emissions by trailing shoes and trailing hoses compared to broadcast spreading was almost identical, 23% and 25% respectively, when applying cattle slurry onto grasslands of four different soil types. Häni et al. (2016) reported that no significant difference in reduction obtained by the two methods was found after application of neither pig nor cattle manure on grassland. The difference in the effect of trailing shoes in experiment H compared to D and E can most likely be assigned to soil conditions, as the slurry properties and air temperatures are similar. The water contents of the soils were in the same range, but the dry bulk density of the soil used in experiment H is much higher than for the soil used in experiment D and E (Table 1). When the soil types and soil water contents are the same, the higher dry bulk density must correspond to a lower amount of air-filled pores. This could hinder infiltration during experiment H compared to the field used in experiment D and E. The main function of trailing shoes is to open the soil surface and expose slurry to porous soil and reduce slurry application to plant foliage. A low soil porosity (high bulk density) is therefore expected to limit the mitigation effect of trailing shoes, since the difference in infiltration rate of shoe versus hose will be diminished.

On average, a significant reduction of 19 ± 12% (paired *t*-test, $p < 0.05$) is found when using trailing shoes compared to trailing hoses during the eight experiments with these two application methods. This agrees with a review by Webb et al. (2010) who found a greater reduction of ammonia volatilization from trailing shoes compared to trailing hoses looking at averages reported in literature. They discuss the large variation in data, which is partly due to the limited amount of reported emissions after application with trailing shoes. Misselbrook et al. (2002) and Wulf et al. (2002) also reported a higher reduction from trailing shoes compared to trailing hoses. However, Misselbrook et al. (2002) only compared the two application methods directly in one out of 27 experiments, whereas in 26 experiments the application methods were compared one by one to broadcast spreading. The other experiments in the study compared either trailing hoses or trailing shoes to broadcast spreading. Contrary to this, Rochette et al. (2008) found significantly higher ammonia volatilization from trailing shoes compared to trailing hoses from pig slurry applied on a clay loam with grass. They assign this unexpected result to a decrease in the infiltration rate caused sealing of the soil surface by the trailing shoes. The variation in reported results can most likely be attributed to factors like soil type, soil and slurry properties, and meteorological conditions, all affecting the infiltration rate of the slurry and thereby the rate of emission. Other challenges also exist, such as the large variation in methods used and the variability of these. Additionally, a large variation in trailing shoe designs exist which has a huge effect on the slits created. The practical implementation of the application method when applying the slurry can also influence results. The majority of articles reports the application of slurry by trailing hoses at the soil surface, however Rodhe et al. (2006) and Smith et al. (2000) reports that the trailing hoses applied slurry 3–10 and 5 cm above ground, respectively, which is assumed to have a significant effect on the emissions.

To avoid tear on the trailing hoses and protect the boom construction it is normal practice in Denmark (observation) to raise the hoses above the crop. In three experiments (H, I and J), all on loamy sand with clover grass, significantly higher ammonia volatilization was observed by using trailing hoses raised 20 cm above grass canopy compared to application at the grass canopy surface ($p < 0.05$). The average increase is 40 ± 13% (Fig. 6). The emission is substantially larger in H compared to I and J. Several factors could cause this. The soil-water content in H is lower than in the other experiments. This could cause lower infiltration rates of the slurry, hence higher emissions as found by Smith et al. (2000). They attribute a lower infiltration to the hydrophobicity of dry soil. These findings are in contrast to findings by de Jonge et al. (2004) and Sommer and Jacobsen (1999) who found that lower soil-water content increased infiltration. The average soil temperature is 5–7 °C higher during experiment H (Table 3), which will cause higher emissions. The lower values of slurry and soil pH during experiment H (Tables 1 and 2), leads to a larger fraction of TAN to be on the ammonium form, which consequently should result in reduced emissions. Conversely, the lower dry matter content of the slurry and higher slurry application rates in I and J could enhance infiltration and thereby lower emissions as observed by Huijsmans et al. (2018) and Häni et al. (2016). This effect should be investigated further in order to draw conclusions.

5. Conclusions

A new system of wind tunnels and online measurements was used to measure the ammonia emissions from band applied liquid slurry. From ten experiments, it was concluded: (i) The wind tunnel set up with online measurements of ammonia allows for precise and repeatable results of ammonia emissions when comparing different treatments or soil types. The online measurements ensure a high time resolution of 104 min, an overall low variation within treatments (coefficient of variation of 13 ± 8%), long measuring times and comprehensive datasets which shows the emissions dynamics over time. (ii) The method for evaluating turbulence intensity in the emission chamber and the effect of turbulence at the soil surface throughout the chamber can be used as a support tool to choose the most realistic air exchange rate. (iii) Trailing shoes were found to give lower ammonia emissions when used on coarse sand, whereas only a weak and varying effect was found for application on loamy sand with and without clover grass and for application on sandy loam. The average reduction of ammonia emissions from trailing shoes compared to trailing hoses was 19 ± 12%. (iv) A high emission reduction can be obtained by correct use of trailing hoses, i.e. by ensuring that the slurry is applied right at the soil surface and hence avoiding splashing.

When choosing the application method with the objective of ammonia abatement, the soil type should be taken into consideration. The results in this study help explain why previous research studies have found different effects of trailing shoes.

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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.

CRediT authorship contribution statement

Johanna M. Pedersen: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft, Visualization. **Anders Feilberg:** Conceptualization, Methodology, Validation, Writing - review & editing, Supervision, Funding acquisition. **Jesper N. Kamp:** Methodology, Writing - review & editing. **Sasha Hafner:** Methodology, Validation, Writing - review & editing, Supervision. **Tavs Nyord:** Conceptualization, Methodology, Validation, Writing - review & editing, Supervision, Project administration, Funding acquisition.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2020.117562.

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