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# Ammonia emission after slurry application to grassland in Switzerland

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# HIGHLIGHTS

• We conducted 17 field experiments on NH<sub>3</sub> emission after slurry application.

- Emissions after slurry broadcast application ranged from 10% to 47% of TAN.
- Examined abatement techniques proofed to be efficient in reducing emissions.

• A regression analysis was performed.

• Air temperature and slurry dry matter were important predictor parameters.

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## ABSTRACT

Loss of ammonia (NH<sub>3</sub>) after field application of livestock slurry contributes between 30% and 50% of agricultural NH<sub>3</sub> emissions from European countries. The objectives of this study were to re-evaluate NH<sub>3</sub> emissions following application of cattle and pig slurry to grassland in Switzerland and to investigate the effectiveness of abatement techniques. In 17 field experiments, NH<sub>3</sub> emissions were determined with a micrometeorological approach, relating the emission to the measured concentration by means of atmospheric dispersion modelling. The cattle slurry applied exhibited an average dry matter content of 3.3% (range between 1.0% and 6.7% dry matter). The emission after application of cattle slurry spread with a splash plate (referred to as reference technique) ranged from 10% to 47% of applied Total Ammoniacal Nitrogen (% of TAN) and averaged to 25% of TAN. This range of losses is lower by approx. a factor of two compared to measurements from earlier Swiss experiments. Applications with trailing hose and trailing shoe systems yielded an average reduction of 51% and 53%, respectively, relative to the reference technique. A regression analysis showed that the dry matter content of the slurry and the air temperature are important drivers for NH<sub>3</sub> emission.

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# 1. Introduction

Ammonia (NH<sub>3</sub>) emissions in Europe are dominated by the agricultural sector (Van Der Hoek, 1998). Field application of livestock slurry is a key source, contributing between 30% and 50% of total emissions in European countries for liquid manure systems (Reidy et al., 2008) which represent more than half of the manure production in most central European countries (Menzi, 2002). National emission inventories in European countries are calculated annually using nitrogen flow models that include the emission

\* Corresponding author. E-mail address: christoph.haeni@bfh.ch (C. Häni). stages of grazing, housing, storage and field application of livestock manure. Emissions of the individual stages are estimated as the product of the nitrogen flow through a stage (e.g. Mg N yr<sup>-1</sup>) and its related emission factor (EF). EFs are fixed individually for the national inventories (Reidy et al., 2008) or based on emission guide-line documents such as the EMEP/EEA air pollutant emission inventory guidebook (EEA, 2013).

Based on an analysis of published data, Sintermann et al. (2012) found a large range of EFs for broadcast application of slurry. Surprisingly, medium plot sizes, typically circles with 40 m diameters, showed systematically higher EFs than larger sized field-plots. Other factors such as air temperature or dry matter (DM) content of the slurry showed no correlation to the emission level. This is in clear contradiction to many other investigations that focused on







specific influencing factors which especially showed a dependency from the DM content, with higher emissions resulting from greater DM contents (e.g. Misselbrook et al., 2005; Sommer and Hutchings, 2001). A large share of the data in Sintermann et al. (2012) that are based on measurements using medium plot sizes originates from the Netherlands (Huijsmans et al., 2001, 2003) and from Switzerland (Menzi et al., 1998). The Dutch experiments covered a DM range from 5% to 15%, while the Swiss data exhibited lower DM contents (1%-7%). Recent measurements in Switzerland, carried out between 2006 and 2010 (Ammann et al., 2012; Sintermann et al., 2011a, 2011b; Spirig et al., 2010), yielded EFs 50% lower than the first generation data from Switzerland. These newer measurements were performed under agronomical conditions similar to the ones used by Menzi et al. (1998) with regard to crop cover, soil properties, timing of application, a DM content of the slurry between 1% and 5% and an average application rate of  $30 \text{ m}^3 \text{ ha}^{-1}$ .

In this paper, we present results from a new series of NH<sub>3</sub> emission measurements at several sites of the Swiss plateau. Since NH<sub>3</sub> emissions after slurry application generally show a rapid decline over the first few hours (e.g. Huijsmans et al., 2001; Spirig et al., 2010) we employed custom-built impinger sampling systems to measure NH<sub>3</sub> concentration with a high time resolution. For the determination of the emissions from the measured NH<sub>3</sub> concentrations, we used a backward Lagrangian stochastic (bLS) dispersion model (Flesch et al., 2004). The primary aim was to reevaluate EFs for slurry application with a focus on spreading of cattle slurry using a splash plate. Additionally, other factors which might influence the emissions were investigated: the experimental plot size, the application technique, the slurry characteristics and the timing of application.

# 2. Material and methods

### 2.1. Field experiments

In total, 17 field experiments were performed between summer 2011 and spring 2014 (Table 1). The field experiments were carried out at five different sites in the Central Plateau of Switzerland (Table 2). At four locations the vegetation cover consisted of a temporary ley and at one site of natural grassland. Application of slurry was done using a tractor pulled tanker.

Slurry was applied onto two to four rectangular plots with standard plot dimensions of approx. 30 m by 30 m. The slurry application duration was between 3 and 10 min per plot and time lags between the slurry applications of different plots were kept as short as possible (approx. 30 min). The areas covered with slurry and the positions of all instruments were recorded by a Global Positioning System (GPS Trimbel R8 GNSS, approx. precision 10 cm). The target application rate for all experiments was  $30 \text{ m}^3 \text{ ha}^{-1}$ , which is the typical application rate in Switzerland. Immediately after application, one NH<sub>3</sub> sampling device (Section 2.2) was positioned at the centre of the plot and the measurement was initiated. The sampling started with short intervals which were then gradually increased as the emissions decreased. The background NH<sub>3</sub> concentration was measured upwind of the experimental area with sampling intervals of 4-8 h. Measurements of wind velocity and background NH<sub>3</sub> concentration were started before slurry application. The duration of emission measurements lasted between 17 and 120 h, but in most cases more than 36 h (Supplementary material I). For each plot, two samples of slurry were taken from the tanker: one before and one after application. The slurry samples were stored at 4 °C until they were analysed in the laboratory. For the determination of the emissions, we used the atmospheric dispersion model 'WindTrax', a bLS model (Flesch

### et al., 2004) (Section 2.3).

Three field experiments (F1 to F3) were carried out to identify potential differences in NH<sub>3</sub> emissions due to the source area size (Table 1). Emissions from standard size plots were compared with emissions from field-scale plots. The dimensions of the field-scale plots were approx. 5000 m<sup>2</sup>. In experiment F1, a third plot with a size of 100 m<sup>2</sup> was investigated. During slurry application on fieldscale plots, it was necessary to refill the slurry tanker twice which delayed the subsequent application operation by roughly 30 min. As a consequence, the field-scale plots could not be treated as homogeneously emitting source areas and the emission rates of the individual tracks were fitted to a bi-exponentially decaying time course assuming identical time behaviour (Sintermann et al., 2011a) for the first few hours, where differences in the emission rates were considerable. Afterwards, the field was treated as a homogeneous emission source. Fourteen field experiments were performed in order to compare application techniques (A1 to A6), slurry characteristics (S1 to S6), and application timing (T1 and T2). Application techniques were splash plate (SP, reference technique), trailing hose (TH), trailing shoe (TS) and shallow injection (SI). Slurry types included slurry from dairy cows (CS) and pig slurry from fattening pigs (PSf) and breeding pigs (PSb). Experiments S4 to S6 were carried out using CS with a DM content between 4.9% and 6.7%, which is higher than the average for slurries used in Switzerland (Flisch et al., 2009).

Additionally in three experiments (T1, W1, W2), the results from the bLS method were compared to results from an independent mass balance method (integrated horizontal flux, IHF) using concentrations and wind measurements at 4 to 6 different heights. One comparison was part of the experiment T1 and two additional comparisons (W1, W2) took place at the site Witzwil. Further information on the applied IHF method is provided in the Supplementary material II.

### 2.2. Automated impinger sampling devices

NH<sub>3</sub> air concentrations were measured using automated impinger sampling devices (low cost impinger systems: LOCI), also known as acid traps. Ambient air was drawn through a threaded midget impinger with a volume of 22 mL (64712-U, Supelco) containing 15 mL of a 0.01 M sodium acetate buffer solution at pH 4. The NH<sub>3</sub> was retained in the acidic solution as ammonium (NH $_4^+$ ). A small amount of dichloromethane was added to prevent decomposition of NH<sup>+</sup><sub>4</sub> by microbial activity. The sampled NH<sup>+</sup><sub>4</sub> concentration was determined in the laboratory by spectrophotometry (DR2800, Hach Lange GmbH). On each LOCI system, seven impinger positions were run subsequently and at two heights (between 0.6 m and 1.6 m above ground level) simultaneously. One additional position was used as a blank probe, exposed but without air flow through the solution. The individual measurement heights are provided in the Supplementary material III. The interval length for each sampling position was set prior to the sampling start on an automation module (Siemens Logic Module) ranging from 10 to 30 min in the first hours after spreading and over 1-6 h during the rest of the time. This automated sampling system guaranteed temporary high resolution sampling without the necessity for onsite control. The air flow through the impinger tubes was kept at approx. 0.7 L/min and logged every 30 s. The NH<sub>3</sub> collection efficiency of the impinger, operated at a flow rate of 0.7 L/min, was higher than 99% for the range of NH<sub>3</sub> concentrations expected to occur in the ambient air.

### 2.3. Emission estimation with a dispersion model

The bLS model that was used for the determination of the

#### Table 1

Summary of the 17 field experiments. ID: identification of the individual experiments. Tech: Application techniques: SP = splash plate, TH = trailing hose, TS = trailing shoe, SI = shallow injection. Slurry types: CS: cattle slurry, PSf: pig slurry from fattening pigs; PSb: breeding pigs.

ID	Date of application	Site	Tech.	Slurry type	Plot size (m <sup>2</sup> )	Application time (CET)	ID	Date of application	Site	Tech.	Slurry type	Plot size (m <sup>2</sup> )	Application time (CET)
F1-Mp	09/20/2011	Tänikon	SP	CS	140	10:10	A6-Th	05/06/2013	Posieux	TH	CS	1590	10:30
F1-Sp					1030	10:20	A6-Ts			TS		1120	12:35
F1-Fp					5350	10:40	S1–Cs	03/15/2012	Wengi	SP	CS	1080	12:55
F2-Fp	11/08/2011	Posieux	SP	CS	6030	11:15	S1-Psb				PSb	1080	13:45
F2-Sp					990	12:40	S1-Psf				PSf	1060	14:30
F3-Sp	05/10/2012	Hohenrain	TH	CS	1330	08:40	S2-Psb	05/24/2012	Wengi	SP	PSb	1080	08:40
F3-Fp					5080	09:10	S2–Cs				CS	1140	09:15
A1-Sp	07/14/2011	Tänikon <sup>a</sup>	SP	CS	1420	10:30	S2-Psf				PSf	970	09:55
A1-Th1			TH		1470	11:00	S3-Psb	11/19/2012	Wengi	SP	PSb	920	11:30
A1-Th2			TH		1360	11:20	S3-Psf				PSf	1010	12:15
A2-Ts	03/12/2012	Posieux	TS	CS	1160	11:15	S3–Cs				CS	930	12:55
A2-Sp			SP		1230	11:30	S4-1	06/12/2013	Wengi <sup>a</sup>	SP	CS	1020	09:50
A2-Th			TH		2080	12:25	S4-2					890	10:20
A3-Ts	07/04/2012	Posieux	TS	CS	980	09:55	S4-3					970	10:55
A3-Th			TH		1480	10:10	S5-Sp	07/10/2013	Wengi	SP	CS	870	11:00
A3-Sp			SP		970	10:35	S6-1	03/11/2014	Wengi	SP	CS	1060	10:20
A4-Ts	08/14/2012	Posieux	TS	CS	950	09:40	S6-2					1080	10:55
A4-Th			TH		1370	10:10	S6-3					980	11:30
A4-Sp			SP		1040	10:30	T1-Ev	08/27/2012	Tänikon	SP	CS	1550	17:15
A4-Si			SI		1090	13:05	T1-Mo	and				1590	06:05
A5-Ts	11/14/2012	Posieux	TS	CS	950	11:25	T1-No	08/28/2012				1520	10:35
A5-Th			TH		1370	11:40	T2-Ev	07/04/2013	Tänikon	TH	CS	990	17:10
A5-Sp			SP		1040	12:00	T2-Mo	and				930	06:40
A6-Sp	05/06/2013	Posieux	SP	CS	1030	10:10	T2-No	07/05/2013				940	12:15

<sup>a</sup> Other influencing parameters varied in the experiments: canopy height: A1-Sp, A1-Th1 A1-Th2; slurry DM content: S4-1, S4-2, S4-3; application rate: S4-1, S4-2, S4-3. More information is provided in the Supplementary material I.

emissions in all 17 field experiments is described in detail by Flesch et al. (2004). The model is incorporated in the WindTrax software (http://www.thunderbeachscientific.com; version 2.0.8.8) in the form of the inverse mode (backward in time) option. It calculates the concentration to emission ratio of finite homogeneous source areas embedded in an area with no biosphere-atmosphere exchange. In order to obtain a unique solution for all present source areas, at least one concentration measurement within each emission plume must be available.

The following input parameters are required to run the model: the friction velocity  $u_*$  (m s<sup>-1</sup>), the roughness length  $z_0$  (m), the Obukhov length L (m), the standard deviation of the rotated wind components  $\sigma u$ ,  $\sigma v$ ,  $\sigma w$  (m s<sup>-1</sup>), the average wind direction and the geometric setup of the source area(s) and the concentration sensor(s). The input parameters were calculated as 10 min averages from sonic anemometer measurements (WindMaster<sup>TM</sup>Pro, Gill Instruments Limited, Lymington, UK). Additional information on the calculation of the input parameters is given in the supplementary material IV.

The modelled average concentration-to-emission rate ratio  $(CE_{bLS})$  was calculated for each sensor and each source area individually on the 10 min intervals and then averaged to fit the actual NH<sub>3</sub> concentration sampling intervals (Section 2.2). As the NH<sub>3</sub> sampling time was adapted to the expected emission level, as well

as to the presumed stability regimes, the resulting averaging error was minimized. The average emission rate (E) for a sampling interval is given as

$$\mathbf{E} = \left( \mathbf{C}_{\text{center}} - \mathbf{C}_{\text{inflow}} \right) / \mathbf{C} \mathbf{E}_{\text{bLS}} \tag{1}$$

where  $C_{center}$  represents the measured NH<sub>3</sub> concentration at the centre of the plot and  $C_{inflow}$  the superposition of the measured upwind background concentration and the concentration interference from the other experimental plots. The concentration measurements from a height of approx. 1 m were taken to calculate the emission rates. The other height was used as a quality check and for gap filling.

### 2.4. Regression analysis

A regression analysis was performed based on the current data set to reveal the most important NH<sub>3</sub> emission drivers and to estimate their effects. The analysis was limited to data from the reference system: splash plate/cattle slurry. The response variable is given by the cumulative NH<sub>3</sub> emission measured during the first 24 h after application in % of applied Total Ammoniacal Nitrogen (% of TAN), denoted as *Loss24h*:

Table 2

Characterization of the experimental sites of this study: farming type, soil texture class, plant cover, geographic coordinates and altitude.

Site	Farming type	Plant cover	Lat./Long. (WGS 84)	Altitude (m.a.s.l)	Soil texture class	Soil pH
Hohenrain	Combined livestock-arable farm	Natural grassland	47°10′57″N/8°19′27″E	600	Loam	6.5
Posieux	Livestock- farm	Temporary ley	46°46'20"N/7°06'07"E	665	Loam	6.6
Tänikon	Combined livestock-arable farm	Temporary ley	47°28′47″N/8°54′17″E	540	Clay	6.7
Wengi	Combined livestock-arable farm	Temporary ley	47°05'30"N/7°24'29"E	475	Clay loam	7.2
Witzwil <sup>a</sup>	Combined livestock-arable farm	Temporary ley	46°59'06"N/7°04'36"E	430	Sandy loam	n.d.

<sup>a</sup> Two additional experiments (W1 and W2) on the comparison of bLS approach with the IHF method; n.d. not determined.

$$\text{Loss24h} = \frac{\text{N24h}}{\text{TAN} * \text{AR}} * 100$$
(2)

where *N24h* is the measured, cumulative loss of NH<sub>3</sub> in kg NH<sub>3</sub>–N ha<sup>-1</sup>, *TAN* is the TAN content in g N kg<sup>-1</sup> of the applied slurry and *AR* is the application rate of the slurry in t ha<sup>-1</sup>. Since *Loss24h* is limited to positive values, a log-transformation of *Loss24h* was applied, where the predictor variables are linear in the transformed response variable:

$$ln(Y) = \beta_0 + \sum \beta_i X_i + \epsilon$$
(3)

This transformation introduces an interaction between the individual predictor parameters, i.e. the effect of a change in individual predictor parameters depends on the level of the response variable. The following predictors were investigated: the weighted (Section 3.2) ambient air temperature (*TAir*) and wind speed (*WS*), the canopy height (*Canopy*) and the dry matter content of the slurry (*DM*).

The regression analysis was done with the statistics software R (R Core Team, 2014) using the least squares routine *lm* for general linear models. The model selection was based on the second-order Akaike information criterion (AICc) based on the method as discussed in details by Burnham and Anderson (2002). The model selection calculations were done using the R packages *AICcmodavg* (Mazerolle, 2014) and *MuMIn* (Barton, 2014). A model set based on all possible combinations of predictor parameters was examined. The parameters that provided the highest predicting capabilities were further averaged over the examined model set. The parameter averaging was performed using weighted averaging over all models that included the predictor parameter, based on the accordant Akaike weights (Burnham and Anderson, 2002).

# 3. Results and discussion

## 3.1. Quality assurance of emission rate determination

### 3.1.1. Reproducibility of single measurements

In experiment S6, the emissions from three application events, which were identical in terms of slurry characteristics, amount of applied slurry, application technique, and plot size were compared to assess the reproducibility of the emission rate determination. Two individual bLS model input parameter sets were calculated from two sonic anemometers that were located at the same height (1.15 m above ground), but at different positions in the field. The results from the two concentration measurement heights (0.6 m and 0.9 m) on each plot yielded a total of 12 emission calculations. The average emission after 48 h was 31.8% of TAN. Individual estimations ranged from 25.4% of TAN to 38.3% of TAN, with an estimated standard deviation of 4.6% of TAN. Therefore, the relative uncertainty of a single NH<sub>3</sub> loss measurement was determined to be  $\pm$  30% (2 $\sigma$ ) of the calculated NH<sub>3</sub> loss. This uncertainty range is in the same order of magnitude as can be expected from a bLS emission estimation as well as from other micro-meteorological methods (Harper et al., 2011).

# 3.1.2. Comparison of the bLS method with the mass balance approach (IHF)

The IHF method (Denmead, 1995) is generally considered robust (Laubach, 2010) and thus often recognized as a reference method. In three experiments (T1, W1, W2), the results obtained by the bLS approach were compared to the IHF method, which is frequently applied in slurry emission studies (see e.g. Huijsmans et al., 2001; Huijsmans et al., 2003). The duration of the measurements was

44 h for T1 and 40 h for both W1 and W2. The results from the comparisons are shown in Fig. 1. The error of the IHF approach of 20% ( $2\sigma$ ) was adopted from Ryden and McNeill (1984) and can be regarded as a lower limit as it does not contain the uncertainty related to the correction of the horizontal diffusion (Supplementary material II). The outcomes agree well and there is no evidence of a systematic difference between the two approaches. Similar results are presented in Harper et al. (2011). It can thus be concluded that the setup of the field experiments in combination with the bLS method provides unbiased emission estimations relative to an IHF approach.

# 3.1.3. Dependency between the source area size and the emission level

The three experiments F1, F2 and F3 did not reveal a systematic higher emission level for medium plot sizes as discussed in Sintermann et al. (2012). Experiment F1 even shows a decreasing emission level trend with smaller plot sizes (Fig. 2). But the discrepancies in each experiment were within the expected uncertainty range of the individual NH<sub>3</sub> emission calculation. Possible biases due to advection between the plots can be excluded because the required corrections due to this effect were smaller than 15%. Thus, the source area size cannot explain the substantially higher EFs from standard size plots compared to field scale plots.

# 3.1.4. Extrapolation of measured NH<sub>3</sub> loss to 96 h

The measured emissions followed a distinct diurnal pattern, with almost no emissions during the night and detectable emissions during the day, peaking around noon. For the extrapolation of the 96 h cumulative  $NH_3$  loss, sequential day-by-day ratios were used. On average, 15% of the first day's emission was observed on the second day, 31% of the second day on the third day and 56% of the third day on the fourth.

The average proportion of the measured NH<sub>3</sub> loss after 24 h to the extrapolated loss is in the range between 80% and 90%. This large fraction of the total loss within the first hours has been observed in other studies before (e.g. Huijsmans et al., 2001; Misselbrook et al., 2002; Pain et al., 1989; Spirig et al., 2010).

### 3.2. Overview on measured NH<sub>3</sub> emissions

Fig. 3 summarizes the cumulative NH<sub>3</sub> losses. The most important experimental parameters and the emission data of each experiment are provided in the Supplementary material I. The average NH<sub>3</sub> emission extrapolated to 96 h after application of cattle slurry with a DM content between 1.0% and 6.7% (average DM content: 3.3%) using the reference technique splash plate amounted to 25% of TAN (range: 10%-47% of TAN). The cumulative emission (96 h) was 14% of TAN (range: 4%–28%) for trailing hose, 9% (range: 4%–12%) for trailing shoe, and 7% for shallow injection systems. Average emissions after 96 h for pig slurries with splash plate application amounted to 11% of TAN (range: 6%-15%), which is lower than the corresponding average emission from cattle slurry (16%). This discrepancy cannot be completely explained, even though the DM content may partly account for the measured difference. It also holds consistent with the results obtained by Søgaard et al. (2002). Overall, these values have to be considered as indicative numbers, which do not allow for a direct comparison between different techniques and slurry types. Nevertheless, the results demonstrate that the emission level obtained in the present experiments is similar to other recent measurements (Sintermann et al., 2011a; Spirig et al., 2010) for cattle slurry, but significantly lower compared to previous studies from Switzerland (Menzi et al., 1998) and values given by UNECE (2014).

The meteorological data (Supplementary material I) represents



**Fig. 1.** Comparison of the two different approaches bLS and IHF. Field experiments: T1 Tänikon Aug 2012, W1 Witzwil Sep 2014 Exp 1, W2 Witzwil Sep 2014 Exp 2. The bars show the average NH<sub>3</sub> loss at the end of the experiment. Lines indicate the approx. uncertainty of bLS  $\pm$  30% (2 $\sigma$ ) and IHF  $\pm$  20% (2 $\sigma$ ).



**Fig. 2.** NH<sub>3</sub> losses in % of applied TAN (at the end of the experiments) from field-scale (FP), standard size (SP) and micro- (MP) plots measured in field trials F1 to F3. Plot sizes were approximately FP: 5000 m<sup>2</sup>, SP: 900 m<sup>2</sup>, MP: 100 m<sup>2</sup>. Vertical lines show ±30% (2σ) of the individual emission calculation.

averaged values over 24 h, using a weighted average that accounted for the typical, observed decay in emission rates. This decay was approximated by a development, based on a Michaelis–Menten type equation (Søgaard et al., 2002) with K<sub>m</sub> (the time at which NH<sub>3</sub> volatilization is 50% of the total loss) equal to 90 min, corresponding to the median K<sub>m</sub> value found for the present study. This weighted averaging guaranteed an improved representation of these influencing parameters under a temporal variation. It increases the weight for the meteorological conditions of the initial period, where the major proportion of the total NH<sub>3</sub> loss occurs (Section 3.1.4).

Fig. 3 shows the cumulative losses versus the application time over the year, stratified by DM content and application technique. Slurries with a high DM content and application during the warm season tend to produce higher losses than slurries with a low DM content and application during the cool season, respectively. The influence of these two factors on the emission level is discussed further in Section 3.4.

Moreover, we compared our results to losses as predicted by the ALFAM model (Søgaard et al., 2002). As shown in Fig. 4, the values of the NH<sub>3</sub> emission extrapolated to 96 h from this study are substantially below the expected range predicted by the ALFAM model. Overall, the dependence of the observed losses on the predicting parameters (air temperature, wind speed, slurry type, DM, and TAN content of the slurry application rate) seems to be captured by the ALFAM model (Fig. 4). However, the stratification of the data according to application techniques and slurry types (Fig. 4 a-c) shows that especially the ones for the trailing hose system are not well reflected.

# 3.3. Effect of NH<sub>3</sub> emission mitigation measures

### 3.3.1. Application technique

The different abatement application techniques have been compared in field experiments A1 to A6. They proved to be efficient in reducing  $NH_3$  emissions. Observed reductions in cumulative

losses at the end of the experiments were on average 51% for trailing hose, 53% for trailing shoe, and 76% for shallow injection systems compared to the splash plate technique (Table 3). For trailing hose application, the decrease in emissions was generally higher than the range given by UNECE (2014), but within the values of Webb et al. (2010). The emission reductions measured for trailing shoe and for shallow injection systems complied well with the numbers published by UNECE (2014).



**Fig. 3.** Measured cumulative NH<sub>3</sub> losses extrapolated to 96 h as percentage of the applied TAN arranged to the time of application over the year and stratified according to the DM content of the slurry: low  $\leq$ 3.5% DM, high >3.5% DM. Acronyms for the application techniques: see caption of Table 1.



**Fig. 4.** Extrapolated cumulative NH<sub>3</sub> losses in % of applied TAN after 96 h vs. predicted total losses from the ALFAM model (Søgaard et al., 2002). Subplots show categories trailing hose/cattle slurry (a), trailing shoe/cattle slurry (b) and splash plate/pig slurry (c) separately. Acronyms for the slurry types and application techniques: see caption of Table 1.

# 3.3.2. Timing of application

Field experiments T1 and T2 suggest that application around noon produced higher emissions than in the morning and in the evening. This is consistent with the findings from previous studies (e.g. Sommer and Olesen, 2000) and can be explained by the higher  $NH_3$  emission potential at noon (Spirig et al., 2010) in combination with the high fraction of total loss occurring soon after application.

## 3.4. Regression analysis of the cumulated NH<sub>3</sub> loss after 24 h

The results of the regression analysis indicate that the ambient air temperature (*TAir*), and the slurry dry matter content (*DM*) are the main drivers in explaining the relative cumulative losses. The remaining predictor parameters wind speed (*WS*) and canopy height (*Canopy*) were found to be of minor importance in the framework of this study. The pH value of the soils of the different field sites are varying within a narrow range and are therefore inappropriate for regression model use. Explanations for exclusion of other parameters from the analyses are given in the Supplementary material V.

The final model is given as:

#### Table 4

Estimates of the regression model coefficients including standard error (SE), lower and upper bound of the 95% confidence interval (CI 95%).

_	Estimate	SE	Lower CI 95%	Upper CI 95%
$egin{array}{c} eta_0 \ eta_{TAir} \ eta_{DM} \end{array}$	1.83	0.16	1.50	2.17
	0.033	0.010	0.013	0.052
	0.18	0.04	0.10	0.25



Fig. 5. Modelled versus observed cumulative  $\mathsf{NH}_3$  losses as fraction of applied TAN after 24 h.

$$\frac{\text{Loss24h}}{[\%\text{TAN}]} = \exp\left(\beta_0 + \beta_{\text{TAir}} * \frac{\text{TAir}}{[^{\circ}\text{C}]} + \beta_{\text{DM}} * \frac{\text{DM}}{[\%]}\right)$$
(4)

with the coefficient estimates averaged over a subset of models that includes only the two predictors *TAir* and *DM* (Table 4). The modelled losses fit the observed losses reasonably well (Fig. 5,  $R^2 = 0.76$ ).

The effect of the air temperature can mainly be related to the correlation between surface and slurry temperatures that control the chemical and physical equilibria of the slurry TAN (e.g. Sommer et al., 2003). Systematic measurements using wind tunnels carried out by Sommer et al. (1991) confirm such an air temperature dependence of the NH<sub>3</sub> loss as well. The DM content of the slurry affects the emissions in several ways (see e.g. Sommer et al., 2003). Higher DM contents inhibit the infiltration into the soil and consequently the sorption of NH<sub>4</sub><sup>+</sup> onto soil surfaces is reduced. Similar to this study, wind tunnel measurements (Misselbrook et al., 2005; Sommer and Olesen, 1991) showed a pronounced

### Table 3

Abatement effect of emission reducing application techniques and values from the literature. n: number of observations.

		Results from this study			Webb et al. (2010)	UNECE (2014)
Application technique Reference		n	Average reduction	Range	Range	Range
Trailing hose	Splash plate	7	51%	[22%, 68%]	[ 0%, 74%]	[30%, 35%] <sup>a</sup>
Trailing shoe	Splash plate	5	53%	[36%, 71%]	[57%, 70%]	[30%, 60%] <sup>a</sup>
Shallow injection	Splash plate	1	76%	-	[60%, 99%]	70%

<sup>a</sup> Higher canopy will increase reduction, depending on placement precision and the extent of herbage contamination (UNECE, 2014).



**Fig. 6.**  $NH_3$  loss in % of TAN of slurry applied to grassland after 24 h arranged by air temperature and DM content of the slurry in percent. Lines: modelled loss; symbols: observed losses.

effect of increasing DM contents resulting in greater NH<sub>3</sub> losses. Fig. 6 illustrates the dependence of the NH<sub>3</sub> loss after 24 h from the air temperature and the slurry DM content when applied onto



Fig. 7. Estimated effect size (incl. 95% CI) of air temperature (TAir) and dry matter content (DM) on the relative NH<sub>3</sub> loss (ALFAM: total loss; this study: loss after 24 h), given a log-linear dependency of the loss (eq. (3)).

grassland as modelled by equation (4) and as observed during the individual emission measurements.

A regression analysis of data from the European database within the ALFAM framework (Søgaard et al., 2002) showed comparable effect sizes for both, the air temperature and the DM content of the slurry (Fig. 7). The more pronounced effect of DM in this study may be attributed to the fact that, for the ALFAM analysis, approx, 20% of the underlying data originates from the old Swiss measurements. These are substantially biased toward too high values and represent data that, regarding the DM content of the slurries, are at the very low end of the data in the ALFAM database. This bias was demonstrated by a thorough check of the raw data still available from Menzi et al. (1998) that contributed to the ALFAM database. It revealed three effects that all contributed to a systematic overestimation of the calculated NH<sub>3</sub> losses: (i) the employed Z<sub>inst</sub> scaling factor (Wilson et al., 1982) is lower by 17%-23% compared to a recalculated value with the current WindTrax version causing a systematic downsizing of the emissions; (ii) a posterior comparison with the data from the nearby (approx. 400 m distance) station of the Swiss Meteorological Service MeteoSwiss showed clear evidence for over speeding of the custom made anemometers during stagnant conditions, especially at night; (iii) the NH<sub>3</sub> advection between the experimental plots which was neglected in the emission calculation of Menzi et al. (1998) can be considerable according to a posterior analysis (Supplementary material VI).

# 4. Conclusions

We performed a new series of NH<sub>3</sub> emission measurements after field application of slurry in Switzerland. The aim was to reevaluate EFs for slurry application as suggested by Sintermann et al. (2012). The focus of the present study was on splash plate application as this is still the reference technique for regulators. The method used for the emission rate determination was thoroughly tested for quality. The uncertainty was in the range which is usually achieved for bLS emission estimation and other micrometeorological methods. A comparison of the bLS method with the IHF approach did not reveal systematic discrepancies. This study found emissions that are systematically lower as compared to earlier Swiss experiments. The Zinst scaling factor, overspeeding of the custom made anemometers and exclusion of NH<sub>3</sub> advection between the experimental plots were identified as factors relevant for the discrepancies. Cumulated emissions predicted by the ALFAM model were higher with an offset of approximately 10%-40% of TAN. A simple regression analysis showed that the DM content of the slurry and the air temperature are important drivers for NH<sub>3</sub> emission. The dependence of the cumulated losses from these parameters is very similar to previously reported values. The emission reductions achieved by the use of trailing hose and trailing shoe applicators were in the order of 50%. Results from the investigation of the application time are in accordance with the diurnal variation of the emission potential.

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### Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.10.069.

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