

Estimating ammonia emission after field application of manure by the integrated horizontal flux method: a comparison of concentration and wind speed profiles

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Abstract

The integrated horizontal flux method is commonly used to estimate ammonia emission from field-applied manure. The method involves measuring the wind speed and ammonia concentration at various heights on a post in the middle of a manured plot. Wind speed and concentration profiles are subsequently fitted to these measurements. The product of the profiles represents the amount of ammonia displaced by the wind, and the calculated ammonia emission is based on integrating the product of the profiles along the height. A crucial step is the functional form of the profiles, and linear relationships employing the logarithm of the height are generally used. In this study, 160 Dutch emission experiments on grassland were re-analysed to evaluate alternative profiles for the concentration and wind speed. It is shown that an exponential concentration model usually provides a better fit than the commonly used profile and that the measurement error for the concentration should be modelled by means of a gamma distribution. Based on the re-analysis, this new model reduces the calculated ammonia emission by around 10%. It is further shown that adding a displacement parameter to the wind speed model only has a minor effect on the calculated emission. Finally, a simulation study reveals that misspecification of the concentration profile may lead to a relative bias of up to 27%, that the precision of the estimated emission can be improved by increasing the number of concentration measurements near the ground and that wind speed measurements at three heights could suffice.

KEYWORDS

displacement height, exponential concentration model, gamma measurement errors, grassland, log wind profile, statistics

1 | INTRODUCTION

Over the last decades, many studies have been carried out to estimate the ammonia (NH_3) emission from field-applied

manure (Hafner et al., 2018). Sintermann et al. (2012) reviewed the various emission measurement methods and their accuracy and uncertainties. A particularly popular method is the integrated horizontal flux (IHF) approach which was

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introduced by Denmead (1983). This method has been applied in a wide variety of studies and countries (Bless, Beinhauer, & Sattelmacher, 1991; Häni, Sintermann, Kupper, Jocher, & Neftel, 2016; Huijsmans, Hol, & Hendriks, 2001; Huijsmans, Hol, & Vermeulen, 2003; Misselbrook, Smith, Johnson, & Pain, 2002; Thompson & Meisinger, 2004). The IHF method was concisely described by Ryden and McNeill (1984) where the horizontal transport of gas 'is determined from the differences in the amount of a gas driven by the wind across the windward and leeward boundaries of an experimental area'. The IHF method as applied by Ryden and McNeill (1984), here referred to as the R&M approach, involves application of manure on a medium-sized plot, measuring the concentration of ammonia at various heights both at a windward location and at a leeward location, and measuring the wind speed at the location. These measurements are used to fit a wind speed and concentration profile by means of a linear regression on the logarithm of the measurement height, from which the resulting ammonia emission is calculated. Goedhart and Huijsmans (2017) noted that, for eight Dutch emission experiments, the concentration profile does not seem to fit the concentration data particularly well and that there is room for improvement. Häni et al. (2016) employed an exponential profile for the concentration without a comparison with the log-concentration model. Furthermore, the IHF wind speed regression model, as applied by R&M, does not include a displacement height parameter which takes into account that, depending on the vegetation or other obstacles on the soil, a zero average wind speed may be reached at some height above the surface (Oke, 1987). Omission of this parameter could exaggerate the horizontal transportation of ammonia close to the ground, resulting in an overestimation of the ammonia emission.

This paper systematically compares the application of different profile functions fitted to concentration and wind speed measurements. The data were obtained for 1191 measurement intervals, or shifts, from 160 emission experiments on grassland which were carried out in the Netherlands between 1997 and 2017. This paper proposes an alternative statistical model for the concentration profile which employs an exponential relationship between the ammonia concentration and height, includes the measured background concentrations to fit the model and uses a gamma distribution for the concentration measurement error. Furthermore, employing the same data, the effect of including a displacement parameter in the wind speed model was evaluated. The resulting emission factors from these modified models were compared with emission factors obtained with the traditional R&M regression model. Furthermore, a simulation study was carried out to determine the bias and precision of the estimate of the emission factor (EF), and to provide advice on the optimal heights at which ammonia concentrations and wind speeds should be measured.

2 | MATERIALS AND METHODS

2.1 | The integrated horizontal flux method to estimate ammonia emission

The IHF method is a micrometeorological mass balance method. It employs profiles of both the wind speed $u(z)$ and the ammonia concentration $c(z)$, measured at the leeward location of an experimental area where manure is applied, as a function of the height z . It is further assumed that the background windward concentration of ammonia, denoted by c_B , does not depend on the height z . The ammonia loss is then given by the difference in flux between the leeward and windward locations:

$$F = \frac{1}{x} \left[\int_{z_0}^{z_p} c(z) u(z) dz - c_B \int_{z_0}^{z_p} u(z) dz \right] \quad (1)$$

The lower integration limit z_0 equals the height at which the wind speed falls to zero, which is given by $u(z_0) = 0$. The upper integration limit z_p is the height at which the concentration of ammonia has decreased to its background windward value c_B , that is $c(z_p) = c_B$. The value of the integral is divided by the fetch of the plot x to give the ammonia emission per unit land area (F).

An important step in the method is the assumed functional form of the profiles $u(z)$ and $c(z)$. Parameters of these profiles are estimated from wind speed and ammonia concentration measurements at various heights. The background concentration c_B is measured at a windward location. Such measurements are typically available for a number of sequential intervals (shifts) after application of the manure. The total emission is then given by the sum of the calculated emissions per shift and further normalized to give the EF expressed as the percentage of emitted $\text{NH}_3\text{-N}$ relative to the total ammoniacal nitrogen (TAN) of the applied manure.

2.2 | The traditional statistical model

Ryden and McNeill (1984) suggested empirical linear relationships for $u(z)$ and $c(z)$ employing the logarithm of the height z : $u(z) = D \ln(z) + E$ and $c(z) = -A \ln(z) + B$. Parameters A , B , D and E are estimated by linear regression, and the integration limits are a function of the estimated parameters: $z_0 = \exp(-E/D)$ and $z_p = \exp((B - c_B)/A)$. Equation (1) can be explicitly solved to give:

$$F = \frac{1}{x} \left[-AD(z \ln^2 z - 2z \ln z + 2z) + (BD - AE - c_B D)(z \ln z - z) + (B - c_B) E z \right]_{z_0}^{z_p} \quad (2)$$

Due to the empirical nature of the R&M model, it may happen that, when using the estimated coefficients A and B , the integration limit z_p is smaller than z_0 or extremely large. This sometimes occurs, mainly in later shifts, when there is little difference between the measured concentration on the plot and the mean background concentration c_B . This results in a flat fitted concentration profile $c(z)$, and the value of z_p is poorly defined. In such cases, fixed values of z_p can be employed, or values derived from a nearby plot or from a previous shift on the same plot.

2.3 | An alternative statistical model for the concentration profile

The R&M concentration model $c(z) = -A \ln(z) + B$ has the flaw that for heights z larger than $\exp(B/A)$, the concentration $c(z)$ is negative, which is clearly impossible. Preferably, for large heights z , the concentration model should have an asymptote which equals the background concentration. A decreasing function with an asymptote for large values of z is given by the exponential model:

$$c^*(z) = \alpha + \beta \exp(-\delta z), \quad (3)$$

in which the α parameter models the background concentration. The exponential model has a physical interpretation since it follows from assuming that the rate of change in the concentration, in the vertical direction, is proportional to the concentration itself with proportionality constant δ . Parameters of the exponential model in Equation (3) can be estimated by using all the concentration data per shift, including the measured background concentrations. This is different from the R&M approach which fits the model employing only concentrations measured at the central post and where the background concentrations are solely used to calculate the upper integration limit z_p . The amount of ammonia on top of the background concentration α is given by $c^*(z) - \alpha = \beta \exp(-\delta z)$, and Equation (1), giving the ammonia loss:

$$F = \frac{1}{x} \int_{z_0}^{\infty} [c^*(z) - \alpha] u(z) dz \quad (4)$$

This employs the same wind profile model $u(z)$ as the R&M model, although the upper integration limit now equals ∞ . In practice, an integration limit z_p such that $\exp(-\delta z_p) = 10^{-6}$ can be used since larger heights will have a negligible contribution to the integral. The integral in Equation (4) cannot be written in explicit form. Instead, numerical integration can be employed. The exponential model for the concentration only

makes sense when both $\beta > 0$ and $\delta > 0$, because only then is a decreasing convex function obtained. Therefore, whenever estimates of β and δ do not comply with this constraint, the estimate of β is set to zero. This results in the constant model $c^*(z) = \alpha$, for which the integral in Equation (4), and thus the ammonia emission, equals zero.

2.4 | Concentration measurement error

The traditional R&M approach employs linear regression to estimate the parameters of the concentration profile which assumes that the measurement error is constant across the profile. However, with measured concentrations between say 50 and 10,000 $\mu\text{g m}^{-3}$, it is more appropriate to assume that the variance of the concentration measurements increases with the mean, such that measurements of higher concentrations are more variable than measurements of smaller concentrations. To derive the distribution of error, such as normal, quasi-Poisson or gamma, the modified Park test for heteroscedasticity (Manning & Mullahy, 2001; Park, 1966) can be used. This test was applied to the residuals and fitted values of the exponential model which was fitted to all shifts (detailed information in Appendix S4).

2.5 | An alternative statistical model for the wind speed profile

The R&M wind profile is given by $u(z) = D \ln(z) + E$. An alternative wind profile, which was fitted to the wind speed data, is given by:

$$u^*(z) = K \ln \left(\frac{z-d}{z_0} \right) \quad (5)$$

This is again a semi-empirical relationship which is commonly used to describe the wind profile in the lowest 20 m of the planetary boundary layer (Oke, 1987). The parameter d is the so-called displacement height, which accounts for vegetation or other obstacles on the soil, while z_0 is the surface roughness. In this model, the wind speed drops to zero at $z = z_0 + d$. The parameter d is often estimated to vary between 0.6 and 0.8 times the height of the canopy (Arya, 1998; Stull, 1988). The wind speed model in Equation (5) simplifies the R&M wind model for $d = 0$, with the R&M parameters $D = K$ and $E = -K \ln(z_0)$.

The traditional R&M wind speed model was compared to two variants of the alternative wind speed model in Equation (5). In the first variant, the displacement parameter d was set to a fixed value equal to $2/3$ of the measured mean height G of the grass, and K and z_0 were estimated from the

wind speed measurements. In the second variant, the parameter d was also estimated. In both cases, a restriction on the parameter space was imposed, such that the height at which the wind speed falls to zero must be equal to or smaller than the grass height G , or $z_0 + d \leq G$. The estimated wind profiles were combined with the R&M and exponential concentration model, and for every combination, the resulting percentage emission was calculated.

2.6 | Ammonia emission experiments and data

In the years 1997–2017, a total of 160 emission experiments were carried out on grassland in the Netherlands. These experiments were run in 11 series where each series has a special purpose such as the direct comparison of two application methods, or the effect of dilution or acidification of manure. Appendix S2 provides the data. The data were used to compare the R&M and exponential concentration models, the wind speed models with and without a displacement parameter, and the emission factors resulting from combining the concentration and wind speed models. Finally, the emission factors, resulting from the exponential concentration model, for the narrowband and shallow injection application methods with untreated manure were compared. This employs quasi-binomial logistic regression, employing a binomial denominator of 100 (McCullagh & Nelder, 1989). The purpose of this analysis was to check whether previously reported significant differences (Goedhart & Huijsmans, 2017; Huijsmans et al., 2001; Huijsmans & Schils, 2009) still hold under the alternative model.

Measurements in the 160 experiments were done for a sequence of four to nine shifts after application of the manure. These shifts were generally short just after manure applications and more prolonged later on. A typical sequence has eight shifts with durations of 1, 2, 3, 3, 15, 24, 24 and 24 hrs, giving four full days in total. The mean wind speed was commonly measured at six heights, typically at 0.28, 0.42, 0.83, 1.38, 2.44 and 3.62 m. For experiments before the year 2005, concentrations were usually measured at seven heights, typically at 0.25, 0.35, 0.57, 0.97, 1.33, 2.01 and 3.32 m, with four background concentrations. Later experiments generally employed five heights (0.25, 0.53, 1.06, 1.98 and 3.29 m) with three background measurements. Further details about the measurement techniques can be found in Huijsmans and Schils (2009). Appendix S1 contains an R (R Core Team, 2018) program to fit the various models to the 160 experiments.

2.7 | Simulation study

A simulation study, employing the exponential concentration model with gamma-distributed measurement errors and the

R&M wind speed model, was performed. This serves four purposes: (a) to determine the precision of the estimate of the emission percentage, (b) to calculate the bias of the R&M model when the true concentration profile is exponential, (c) to see whether the wind speed can be measured at a single height of 1.5 m while assuming that the wind speed falls to zero at a specific height close to the ground and (d) to see how the precision is affected by increasing the number of wind speed or concentration measurements, or by changing the position of heights at which these are measured.

The simulation was based on a single shift of 1 hr with 40 kg ha^{-1} TAN applied to an experimental area with fetch 25 m. The parameters D and E of the R&M wind speed profile $u(z)$ were chosen such that $u(0.03 \text{ m}) = 0$ and $u(4 \text{ m}) = 3.5 \text{ m s}^{-1}$. Normal measurement errors were assumed for the wind speed, and the residual standard error was set to 0.1 m s^{-1} . These parameter values were similar to the mean values of the fitted wind speed profiles over all experiments and shifts. The most crucial parameter in the exponential concentration model is δ because it describes the speed with which the concentration falls to the background level. Therefore, simulations were done for five δ values: 1.0, 1.25, 1.5, 1.75 and 2.0. For these δ values, the concentration drops almost to the background level at a height of 4.6 ($\delta = 1$) to 2.3 m ($\delta = 2$). These heights were obtained by solving $\exp(-\delta \times z) = 0.01$ for z . The corresponding β parameters were chosen such that the resulting emission percentage, given the wind speed simulation model described above, was equal to 15%, that is 6 kg ha^{-1} TAN, for the shift. This resulted in β parameters equal to 1968, 2654, 3402, 4210 and 5076, respectively. The background concentration parameter α was set to 25. Additional simulations were carried out for values of β such that the emission percentage was equal to 10% or to 5%, that is by dividing β by 1.5 or by 3. Concentrations were simulated using gamma distribution with the extra gamma parameter σ^2 set to 0.05 which implies a coefficient of variation of $\sqrt{0.05} = 22\%$. The value 0.05 was obtained by taking the mean of the estimates of σ^2 for every individual shift. The value of 0.05 was more or less constant across shifts and across emission percentages.

In the first three simulations, wind speed and concentrations were simulated at the typical heights which were employed in the 160 experiments. The fourth simulation, which aimed at optimizing the number and locations of measurement heights, employed different numbers of measurements. The measurement heights were chosen to be equidistant on the logarithmic scale between 0.07 and 3.5 m for the wind speed, and between 0.1 and 3.5 m for the concentration. For example, five log-equidistant heights for the concentration are at 0.10, 0.24, 0.59, 1.44 and 3.5 m, while ten log-equidistant heights are given by 0.10, 0.15, 0.22, 0.33, 0.49, 0.72, 1.07, 1.59, 2.36 and 3.5 m. The logarithmic scale was used because both the wind speed and concentration profiles change more rapidly for smaller heights than for larger heights.

For each simulation, 5,000 data sets were generated, and the standard error of the resulting emission percentages relative to the true value, also known as the coefficient of variation CV, was calculated.

3 | RESULTS

3.1 | Results for the Ryden and McNeill model

The R&M profiles for wind speed and concentration were fitted to all 1,191 shifts in all 160 experiments, and Equation (2) was used to calculate the corresponding emission percentage. The concentration profiles were visually inspected, and when the fitted curve was very flat or the profile resulted in an unrealistic large emission percentage, the shift was earmarked as having 'No profile'. A total of 115 profiles (10%) were thus earmarked, mainly in later shifts which usually did not contribute much to the total emission. 'No profile' shifts were excluded from the calculation of the cumulative emission, that is the corresponding emission was set to zero. An example of the data and the fitted profiles for eight shifts in a single experiment is given in Figure 1. Appendix S3 provides such plots for all experiments.

The wind speed profiles in Figure 1 follow the data very closely, and this is the case for most shifts (see Appendix S3). The 10% and 90% quantiles of the residual standard error for all the 1191 fitted wind profiles equal 0.029 and 0.185 m s⁻¹, respectively, while 70% of the values are smaller than 0.1 m s⁻¹ which is the value used in the simulations.

A histogram of the values of z_0 , the height at which the wind profile is zero, is given in Figure 2a; this reveals that most values are smaller than 0.08 m. Figure 2c shows the relationship between the mean of z_0 per experiment and the grass height. Although there is quite some spread in the values of z_0 , a regression line through the origin is highly significant and the estimate of the slope equals 0.39. This suggests that the height at which the wind speed falls to zero can be approximated to 40% of the grass height.

The R&M concentration profiles in Figure 1 seem not to fit very well. For the first six shifts, the curve overestimates ammonia concentrations halfway and clearly underestimates lower concentration values. The R&M method seems to be rescued by the upper integration value z_p , given by the vertical line in Figure 1, which cuts off the negative part of the concentration profile. For the last shift in Figure 1, the value of z_p is larger than 4.0 m. This typically happens when the fitted concentration profile is flat and the mean background concentration is somewhat lower which can happen by chance. A histogram of the values of z_p is given in Figure 2b. Most of the values are between 1 and 3.5 m, although occasionally values larger than 4 m are found. Figure 2d reveals that very

small and very large values of z_p typically occur when the percentage emission in a shift is low, that is when there is hardly a concentration profile.

3.2 | Results for the exponential concentration model

The exponential concentration model with normal errors was fitted to all shifts. The modified Park test for heteroscedasticity resulted in an estimate of 1.87 for the power P in the variance function; detailed results can be found in Appendix S4. This indicates that the measurement errors are more or less gamma-distributed, with $P = 2$, and therefore, the exponential model was fitted employing the gamma distribution.

Fitted exponential profiles, employing gamma measurement errors, are given in Figure 1 and in Appendix S3 for all experiments. The exponential model generally fitted very well. For 29 of the 115 shifts with the 'No profile' earmark, the estimate of β was zero resulting in a constant profile and zero emission in that shift; this can be viewed as an automatic way to earmark shifts with a constant (or 'no') profile. For the remaining 86 earmarked shifts, a statistical test of the hypothesis $H_0: \delta \leq 0$ was not rejected, except for a single case, indicating that the concentration profile was flat or hard to estimate. This supports the earmarking of these shifts. The single exception was shift 8 in experiment 2000-12-05; this shift was earmarked because the previous shift clearly had no profile and because shift 8 in four experiments conducted at the same time also hardly had a profile. Figure 3 displays three unusual cases where only the data in Figure 3b received the 'No profile' label before fitting the profiles. The data in Figure 3a clearly result in a much too large emission percentage. The exponential model in Figure 3b does reasonably well, while the R&M model gives a very large emission. Both emission percentages for the data in Figure 3c are quite large which is due to a single somewhat larger measured ammonia concentration just above the ground. Clearly, a visual inspection of the profiles is required for the exponential model and the R&M model.

The median value of the estimated background concentration parameter α , excluding shifts with a 'No profile' label, equals 21 $\mu\text{g m}^{-3}$. In 3.3% of the cases, the estimated background was larger than 100 $\mu\text{g m}^{-3}$. The median value of the estimates of the exponential rate parameter δ equals 1.9 which implies that at 2.4 m, the concentration almost equals the background value (since $\exp(-1.9 \times 2.4) = 0.01$). In 5.9% of the shifts, the estimate of δ was >5 implying that the background concentration is already reached at 1 m height or less. In 6.1% of the shifts, δ is <1 implying a more prolonged concentration profile, which means that the upper integration limit ∞ in Equation (4), effectively given by z_p such that $\exp(-\delta z_p) = 10^{-6}$, can be quite large.

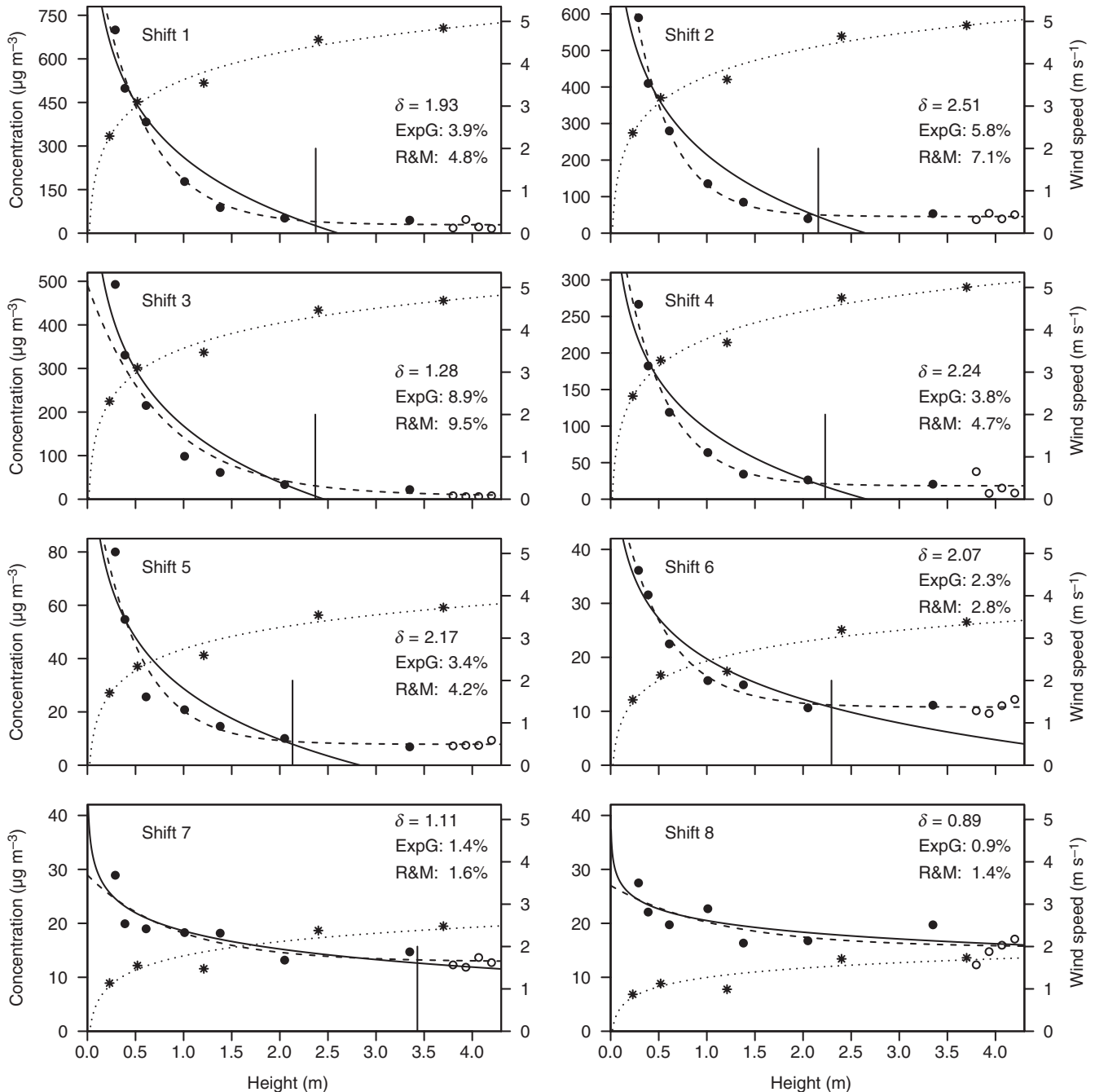


FIGURE 1 Measurements of ammonia concentration (solid circles) and wind speed (stars) at various heights at the central post and measured background concentrations (open circles). The latter are for convenience depicted at a height of around 4 m. The dotted curve is the fitted wind speed profile, the solid curve is the fitted R&M concentration profile, and the dashed curve is the fitted exponential profile with gamma measurement errors. The vertical line denotes the upper integration limit z_p which is used in the R&M model. Each panel displays the estimate of the exponential rate parameter δ and the estimated percentage emission according to the exponential and the R&M concentration profile. The data are for experiment number 56

Ammonia emission at heights above 4 m might be considered unrealistic, and therefore, the integration interval in Equation (4) was split into two parts whenever $z_p > 4$: (a) from z_0 to 4 and (b) from 4 to z_p . For 92% of the 160 experiments, the second integration interval amounts to <1% total emission in absolute terms. This reveals that for most experiments, integration to ∞ is more or less equivalent to

integration up to 4 m. However, for some experiments (notably experiments 2, 19, 25 and 137) the second integration interval gives a large emission in absolute terms (respectively, 5%, 4%, 4% and 7%). The graphs in Appendix S3 show that for these four experiments, the exponential model does not fit too well and that the R&M concentration model might be preferred.

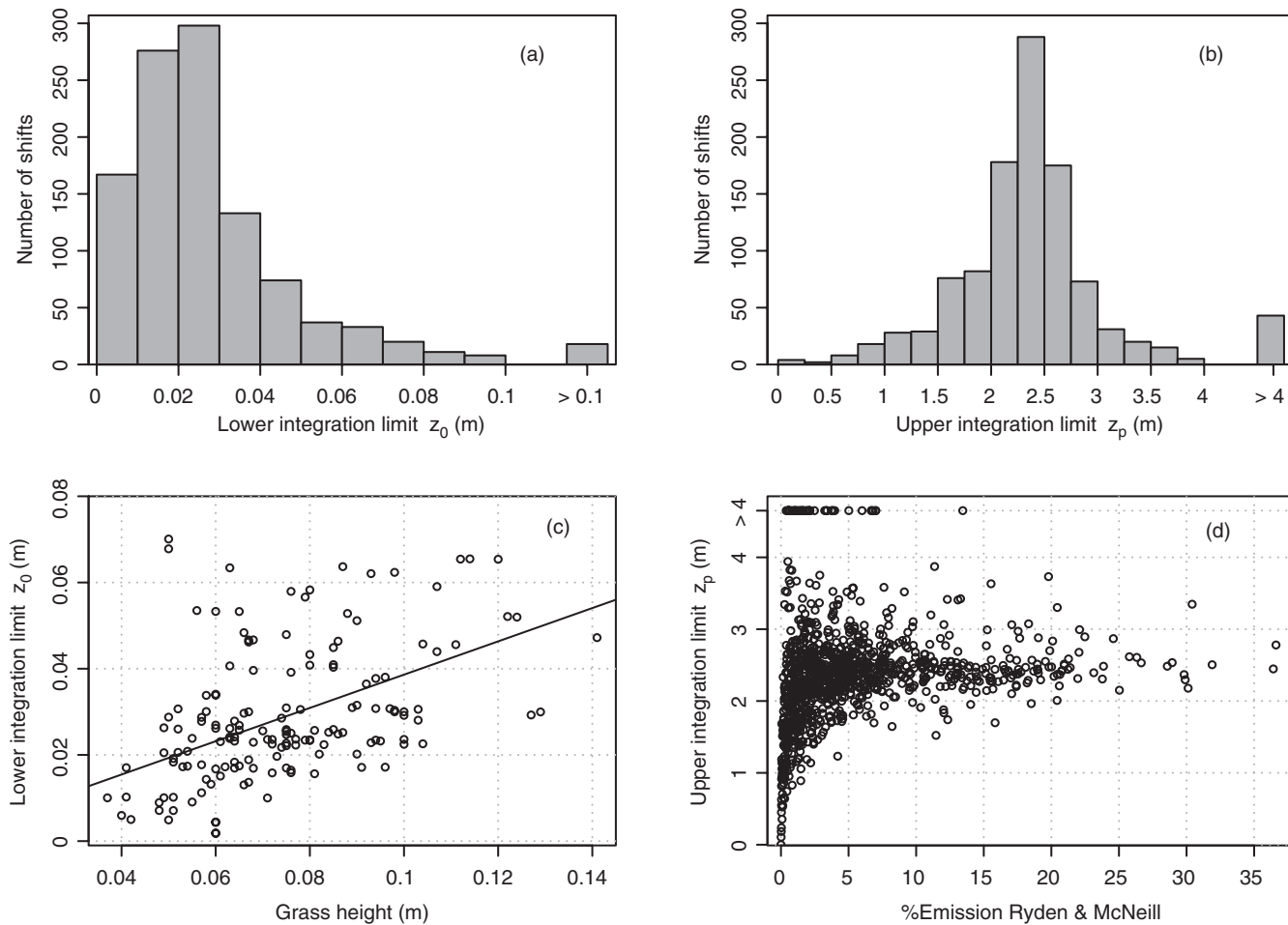


FIGURE 2 Histogram of the integration limits z_0 (the estimate of the height at which the wind speed falls to zero, panel a) and z_p (the estimate of the height at which the mean of the background concentrations is reached, panel b) resulting from the R&M model per shift, only for those shifts where a profile was fitted. Panel (c) displays the mean of the z_0 values per experiment versus the grass height with a fitted line through the origin. Panel (d) depicts the height z_p versus the corresponding percentage emission according to the R&M model for those shifts where a profile was fitted

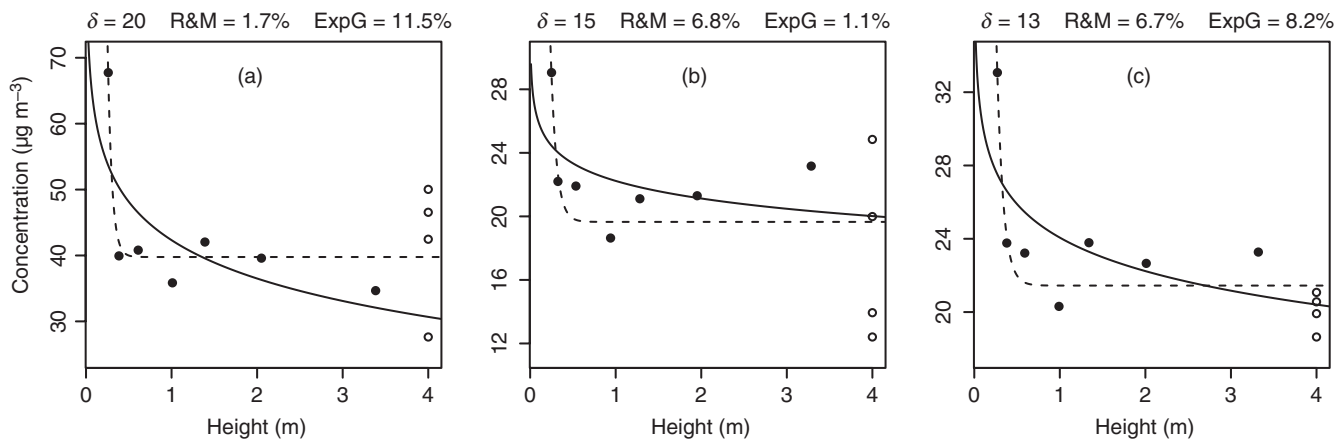


FIGURE 3 Three shifts with unusual concentration profiles. Measured ammonia concentration (solid circles) at various heights at the central post and measured background concentrations (open circles) for shift 8 in experiment 15 (panel a), experiment 33 (panel b) and experiment 85 (panel c). The backgrounds are plotted at a height of 4 m. The solid curve is the fitted R&M concentration profile, and the dashed curve is the fitted exponential profile with gamma measurement errors. The title of each plot gives the exponential rate parameter δ and the emission percentages for the R&M and the exponential (ExpG) model

3.3 | Comparison of total cumulative emissions

Cumulative emission percentages were calculated for the R&M and the exponential concentration model, both combined with the R&M wind speed model. The emission for shifts with the 'No profile' earmark was set to zero, and this was also done for the shift displayed in Figure 3a. Figure 4 displays the cumulative emission percentage resulting from the exponential model versus the percentage resulting from the R&M model, based on all shifts and based on the first five shifts only, that is roughly the first 24 hrs after manure application. The latter is also displayed because the concentration model does not always fit well for later shifts with low emissions. For 89% of the experiments, the exponential model gives a lower cumulative emission percentage than the R&M model. The mean ratio equals 0.90, with 10% and 90% quantiles equal to 0.77 and 1.01, both for all shifts and also for the first five shifts (with similar quantiles). For experiments with a R&M cumulative emission percentage larger than 50%, the mean ratio equals 0.91 with quantiles 0.84 and 0.99. Excluding experiments with a cumulative emission smaller than 10%, the mean ratio for narrowband equals 0.86, for shallow injection 0.89 and for broadcast spreading 0.93. The differences between these ratios are significant with $p < 0.05$. These latter mean ratios can be combined with mean percentage ammonia emission in 199 experiments in the Netherlands (Table 2 in Goedhart & Huijsmans, 2017) to give reduction factors relative to broadcast spreading, when applying the exponential model. This results in a reduction factor of 68% for narrowband and of 80% for shallow injection. This is similar to the reduction factors, of 65% and 78%, obtained using the R&M model.

Figure 5 displays the cumulative percentage emission for the two low-emission techniques shallow injection and narrowband (trailing shoe) application after 24, 48 and 72 hrs, with a margin of 2 hrs. The margin of 2 hrs was used because of different timings of shifts in the various experiments. Only experiments with untreated manure and a shift ending in the specified time slot were used. It is evident that, on average, narrowband application results in higher emission than shallow injection. Table 1 lists the number of experiments and the mean emission percentages for these experiments. The difference between the two application methods, employing quasi-binomial logistic regression, is very significant with p -values < 0.002 for the three chosen periods.

3.4 | Results for two alternative wind speed models

Figure 6 compares the cumulative percentage emission resulting from the R&M wind speed model and the two

alternative log wind speed models which employ a displacement height parameter, when combined with the exponential concentration model. There is hardly any difference between the calculated emissions resulting from the two alternative wind speed models. For most shifts, the lower integration limit $d+z_0$ of the two alternative wind models is larger than the lower integration limit z_0 of the R&M wind model (detailed results are given in Appendix S5). However, in many cases the fitted profiles for larger heights are quite similar. Moreover, even if wind profiles are different for larger heights, the effect on the emission will be small due to much lower ammonia concentrations at larger heights. It follows that the main impact of employing an alternative wind speed model is a smaller integration interval obviously resulting in smaller emissions. On average, the reduction in cumulative emission is $< 3\%$.

3.5 | Results of the simulation study

In the first simulation, the precision of the estimated emission percentage was determined. With wind speeds measured at six heights and four background concentrations, there was no bias and the coefficient of variation CV of the simulated emissions was around 10% when concentrations were measured at 7 heights and around 12% when measured at 5 heights. The CV value did depend on the true value of the rate parameter δ with somewhat larger CVs for larger values of δ . Slightly larger CV values were observed when the β parameter values were divided by 1.5 or by 3.

In the second simulation, the R&M model was fitted to the exponentially simulated concentration values, again with wind speeds measured at 6 heights and 4 background concentrations. The bias was negligible for $\delta = 1$. However, for $\delta = 2$, the emission was overestimated by 20% of the true value when the concentration was measured at 7 heights and 27% when measured at 5 heights. For the intermediate value $\delta = 1.5$, emission was overestimated by 10% and 14%, respectively. So misspecification of the concentration model may lead to a large positive bias especially for large values of δ and when concentrations are measured at a limited number of heights.

In the third simulation, the wind speed was only measured at a single height of 1.5 m and it was assumed that the height at which the wind speeds falls to zero, that is z_0 , equals 0.03 m which is according to the true wind speed model. In that case, there was no bias. However, when z_0 was assumed to be 0.06 the bias ranged from -5% ($\delta = 1$) to -12% ($\delta = 2$). The bias is negative, that is the emission is underestimated, because the part between 0.03 m and 0.06 m is missing in the integral. When the true value equals $z_0 = 0.06$ m and a value of 0.03 m was assumed, the bias ranged from 6% ($\delta = 1$) to 14% ($\delta = 2$). The bias

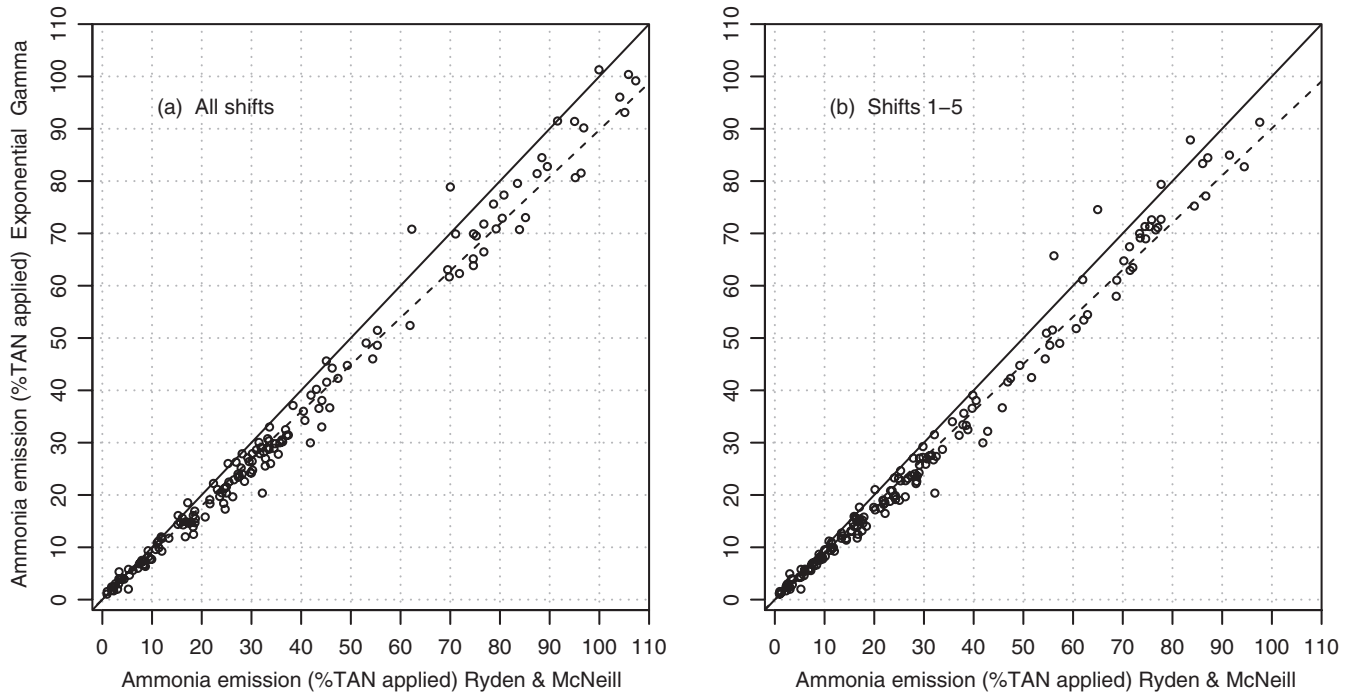


FIGURE 4 Cumulative ammonia emission as a percentage of TAN applied for the 160 experiments resulting from the exponential concentration model with gamma measurement errors versus those resulting from the R&M model, for all measured shifts (panel a) and for the first 5 shifts only (panel b). Both concentration models were combined with the R&M wind speed model. The solid line represents $Y = X$. The dashed line has a regression coefficient of 0.90, that is a 10% difference, and is obtained by means of a generalized linear regression model through the origin employing a Poisson distribution

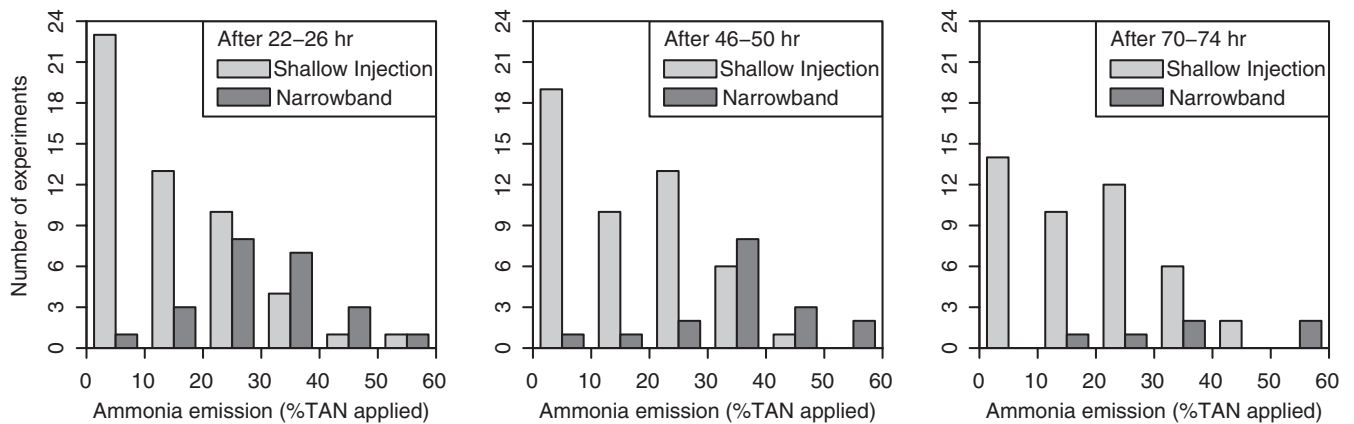


FIGURE 5 Histograms of cumulative ammonia emission as a percentage of TAN applied for shallow injection and narrowband with untreated manure after 24, 48 and 72 hrs with a margin of 2 hrs. The percentages were calculated with the exponential model with gamma measurement errors and the R&M wind speed model

did not depend on whether there are 5 or 7 concentration measurements.

In the fourth simulation, increasing the number of background concentrations from 1 to 16 had little effect on the precision of the estimate of ammonia emission. At most the precision was improved by 5%. However, replacing a single background concentration with an extra concentration measurement at the central post gave a worse precision for

values of $\delta = 1$ and 1.25. This implies that at least a single background measurement is required when the concentration profile declines rapidly. Increasing the number of wind speed measurements only had a minor positive effect on the precision. For example with 5 concentration measurements and $\delta = 1$, the CVs for 2, 3, 4 and 10 wind speed measurements equalled 12.5, 12.2, 12.1 and 12.1%, respectively. With 10 concentration measurements, these CV values were 9.2, 9.0,

TABLE 1 Summary statistics for the ammonia emission as a percentage of TAN applied resulting from the exponential concentration model with gamma measurement errors and the R&M wind speed model, for experiments with non-treated manure in the Netherlands

Application method	After 22–26 hrs		After 46–50 hrs		After 70–74 hrs	
	Count	Mean (SD)	Count	Mean (SD)	Count	Mean (SD)
Shallow Injection	52	13.6 (11)	49	14.0 (10)	44	16.2 (11)
Narrowband	23	26.1 (10)	17	31.2 (11)	6	34.3 (14)

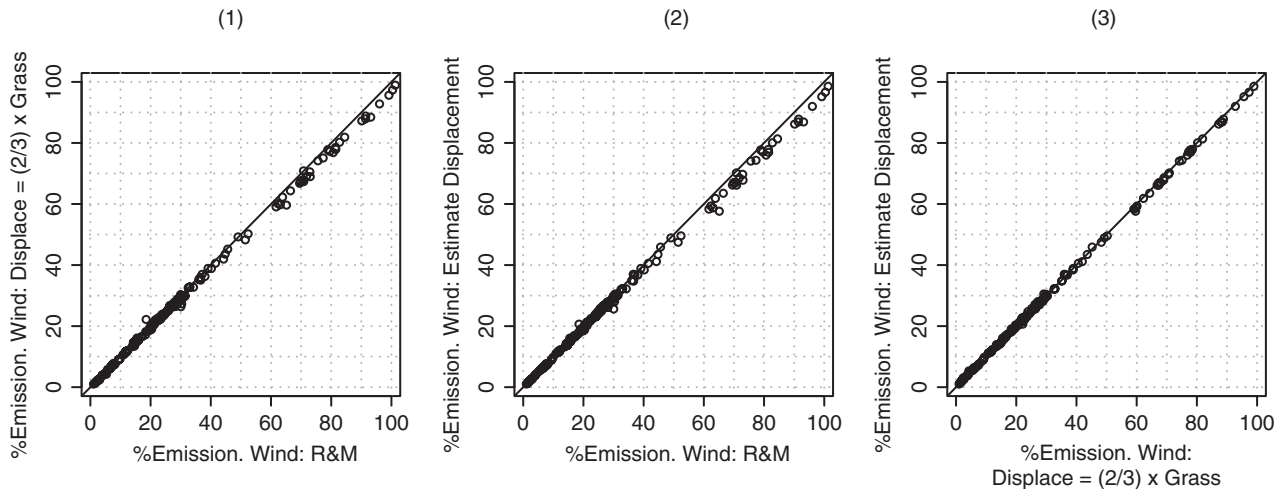


FIGURE 6 Comparison of cumulative ammonia emission percentages for the 160 experiments when employing three different wind speed models combined with the exponential concentration model with gamma measurement errors. The three wind speed models are (1) the traditional R&M log wind model, (2) the log wind model with displacement d set to $2/3$ of the grass height and (3) the log wind model where the displacement parameter d is estimated. The solid line represents $Y = X$

9.0 and 8.6%. The largest increase in precision resulted from increasing the number of heights at which the concentrations were measured at the central post (Table 2). The effect is most prominent for $\delta = 2$ because the decline in concentration is steepest for this value and having extra observations for lower heights is then especially valuable. Finally, the positioning of 10 heights at which the concentration is measured was varied to see whether, for example, having multiple observations at 0.1 m or at 0.24 m was beneficial. The best configuration, among those tested, was the one in which the 10 heights were equidistant on the logarithmic scale, rather than having multiple observations at fewer heights.

The coefficient of variation, as estimated in the first simulation, will largely depend on the size of the measurement error of wind speed and concentration. This was confirmed by a small simulation study, in which 500 data sets were generated per setting, employing gamma measurement errors of the concentration with σ^2 set to 0.10 instead of 0.05. The CV value of the simulated emissions was then around 15% when concentrations were measured at seven heights and around 18% when measured at five heights. The reported bias for the second, third and fourth simulation was not affected by the larger value of σ^2 .

4 | DISCUSSION

It is evident that the concentration model of Ryden and McNeill (1984) does not fit the measured ammonia concentration data particularly well. This is especially the case for concentrations observed at larger heights. Moreover, the measured background concentrations are not used in fitting the R&M concentration model, although they provide information about the concentration at an infinite height. It is therefore proposed to employ the exponential model for the concentrations and to include the measured background concentrations when fitting this model. However, for some experiments the exponential model does not fit too well which may result in relatively substantial emission at heights above 4 m. In such cases, one might prefer to use the traditional R&M model. Häni et al. (2016) also employed an exponential model for the ammonia concentration although they did not include the background parameter α , such that, according to their model, the concentration equals zero for large heights. For the data employed in this paper, with sometimes high background concentrations, a background concentration parameter was necessary.

TABLE 2 Coefficient of variation obtained by simulation expressed as a percentage of the true emission for 5–10 concentration measurements, equidistant on the logarithm scale between 0.1 and 3.5 m, at the central post for five different values of the exponential rate parameter δ . The simulation employed five wind speed measurements, equidistant on the logarithmic scale between 0.07 and 3.5 m, and a single background concentration. See text for simulation parameters

Number of concentrations	$\delta = 1.00$	$\delta = 1.25$	$\delta = 1.50$	$\delta = 1.75$	$\delta = 2.00$
5	12.19	12.09	12.37	12.40	12.49
6	11.25	11.04	11.07	10.92	10.81
7	10.44	10.14	10.15	10.07	10.07
8	9.66	9.49	9.42	9.21	9.38
9	9.15	8.97	8.97	8.94	8.99
10	8.78	8.54	8.55	8.42	8.45

Measured ammonia concentrations may range from 50 to 10,000 $\mu\text{g m}^{-3}$ in a single shift. It is unlikely that measurements with such a broad range have a constant measurement error. This was confirmed by application of the modified Park test for variance heterogeneity to the residuals and fitted values of the exponential concentration model with normal errors. A good alternative appears to be gamma-distributed errors for which the standard error is proportional to the mean. This is in line with many studies which support the use of the lognormal distribution, which is very similar to the gamma distribution, for pollutant concentrations in the air, see, for example, Kahn (1973) and Ott (1990). Gamma measurement errors are therefore proposed.

The alternative log wind profile, with a displacement parameter to accommodate vegetation or other obstacles on the ground, results in emission percentages which are up to 3% lower than with the R&M wind speed model. This is mainly due to a somewhat larger lower integration limit in the IHF integral in Equation (1). The relatively small difference might be due to a low grass height in the 160 experiments and to constraining the lower integration limit to be lower than the grass height. For application of manure in crops with a higher canopy, the effect of including the displacement height may be larger. However, with 'only' six wind speed measurements, it is hard to discriminate between the different wind profiles. So, one might argue that, for the 160 experiments discussed in this paper, the choice of a particular wind speed model is a matter of taste. This does, however, not imply that grass height is unimportant; a higher canopy has a reducing effect on the emission in case of narrowband and shallow injection (Huijsmans et al., 2001; Huijsmans, Vermeulen, Hol, & Goedhart, 2018; Thorman, Hansen, Misselbrook, & Sommer, 2008).

A comparison between the traditional R&M model and the new model (i.e. the exponential model with gamma errors combined with the R&M wind speed model) for the 160 ammonia experiments revealed that for 89% of the experiments, the new model results in a lower emission percentage.

The mean ratio of the two percentages equals 0.90. With the new model, the EF for shallow injection is significantly lower than the EF for narrowband application. This is in line with previously obtained results (Goedhart & Huijsmans, 2017; Huijsmans et al., 2001; Huijsmans & Schils, 2009).

The number of concentration measurements, and their position, varies between IHF studies. Bless et al. (1991) measured at four heights (0.25, 0.65, 1.6 and 4 m), Häni et al. (2016) employed four heights (0.61, 0.9, 1.6 and 3.0 m), Misselbrook et al. (2002) used five heights (0.25, 0.65, 1.2, 2 and 3.3 m), while Thompson and Meisinger (2004) measured the concentration at six heights (0.2, 0.4, 0.9, 1.4, 2 and 3 m). In the 160 experiments described in this paper, the concentration was generally measured at five (0.25, 0.53, 1.06, 1.98 and 3.29 m) or seven heights (0.25, 0.35, 0.57, 0.97, 1.33, 2.01 and 3.32 m). In general, it is best to have more measurements at heights where the concentration profile changes most, that is close to the ground. In the simulation study, the first measurement was therefore positioned at 0.1 m, and further heights were chosen on an equidistant logarithmic scale giving relatively many observations close to the ground. The maximal height at which the concentration should be measured depends on the rate of decline, that is the δ parameter in the exponential model.

A simulation study, employing the exponential concentration model with gamma errors and the traditional R&M wind speed model revealed that the standard error of the estimated emissions, relative to the true emission, was around 10%. This would imply that a simple confidence interval for the estimated percentage equals $\pm 20\%$ of the percentage itself. This is similar to the values between 13% and 37% which were found using a bootstrap approach for eight emission experiments (Goedhart & Huijsmans, 2017). The simulation study also showed that (a) measuring the wind speed at a single height, while assuming that the wind speed falls to zero at a particular height, may result in a large bias whenever the assumed height with zero wind speed is misspecified; (b) that having more than three heights at which the wind speed is

measured has a minor positive effect on the precision which is due to the small measurement error of the wind speed; (c) that a single background concentration suffices when the exponential concentration model is used; and (d) that the largest increase in precision results from increasing the number of concentration measurements at the central post and positioning these equidistant on the logarithmic scale.

Automatic calculation of EFs is discouraged. Every experiment and every shift requires careful examination of the data and the fitted profiles, and an argued choice of the profiles to be fitted. It is essential to earmark shifts as having a 'no concentration profile', setting the corresponding emission to zero. In this study, it was, mainly later shifts, which usually do not contribute much to total emission, that were earmarked. The earmarking was confirmed by testing whether the exponential decay parameter δ of the concentration model is less than or equal to zero. While earmarking remains a subjective choice, it is important to exclude shifts for which the models do not work.

The main Equation (1) to calculate ammonia emission only deals with the horizontal transport of ammonia and lacks a horizontal diffusion, or turbulence, term. Estimation of turbulence requires fast response instrumentation at many heights (Denmead, 1983) rather than time-integrated measurements at relatively few heights. This is why the turbulence component is almost never measured. Instead, the effect of turbulence can be accounted for by applying a general correction term. Several studies have been carried out to quantify this effect. Wilson and Shum (1992) showed that, for plots with a radius of 20 m and a roughness length z_0 of 0.01 m, neglecting turbulence overestimates emissions by a factor of 1.03–1.07, depending on whether conditions are stable or unstable. For a roughness length of 0.1 m, they obtained a factor of 1.10–1.21. Leuning, Freney, Denmead, and Simpson (1985), Wilson, Flesch, and Harper (2001), Desjardins et al. (2004) and Gao, Desjardins, and Flesch (2009) found overestimation factors of 1.05–1.10. Häni et al. (2016) corrected their measurements, in which manure was applied to grassland, by 7% following Wilson and Shum (1992). Equation (1) also employs time-integrated measurements of wind speed and concentration thereby ignoring possible correlations in time between the two. Such time correlations might especially be present in longer measurement periods such as the 24-hr shifts in the experiments discussed in this paper. The effect of time correlation over prolonged shifts is currently unknown. We are working on quantifying this effect.

5 | CONCLUSIONS

Sintermann et al. (2012) noted that the IHF method is 'widely considered a very robust approach'. However, this paper

shows that several choices need to be made when applying the IHF method, such as the length and number of shifts, the number of heights at which the wind speed and concentration are measured, the wind speed and concentration profiles to be used and the decision as to whether there is a concentration profile or not. Moreover, the simulation study reveals that the relative standard error of the estimated emission percentage could be 10%. One could of course allocate more resources to reduce this relative standard error. However, the object of ammonia emission studies is often to compare different application methods or different manure treatments. It is thus more beneficial to repeat experiments, for example under different weather and soil conditions, rather than getting a very precise estimate of the emission for a single experiment which is conducted under specific circumstances. Moreover, any comparative statistical analysis of emission percentages obtained in multiple experiments implicitly takes account of the measurement error per individual experiment (Goedhart & Huijsmans, 2017). Finally, a comparison of, for example, different application methods is best done in a pairwise experiment such that weather, manure and field conditions are similar.

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