Accurate estimation of ammonia emissions after manure application: an interdisciplinary approach

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This work is dedicated to my great-grandfathers Bastiaan Oskam and Otto de Leeuw, during their lives dairy farmers in Lopikerkapel

Abstract
Ammonia (NH_3) emission after manure application to farmland contributes significantly to anthropogenic ammonia emission and its potentially harmful effects on the environment. Internationally, this results in the application of increasingly strict environmental policies. Recent studies however suggest that measured emissions overestimate actual emissions. Emissions can be estimated more accurately when differences in emission between manure application methods are compared with differences in nitrogen (N) recovery from the applied manure by crops. Because N not volatilized as NH_3 is largely available for crop uptake, the results of this comparison indicate the level of misestimation (if present) and can be used for more accurate estimation. This approach is demonstrated for the case of application of liquid cattle manure by surface-spreading (SS) or shallow injection (SI) on permanent grassland grown in a temperate climate. Using data of most recent multi-year field experiments, the difference between SS and SI in recovered manure N is estimated at on average 18 percentage points (pp.). This translates into a difference in manure NH_3 emission factor (EF) of 37 pp. In case of the Netherlands, a country with mandatory SI to reduce NH_3 emissions, the difference between current official EF's for SS (74%) and SI (19%) is 55 pp. Comparison of these differences suggests that official EF's overestimate actual emissions by on average 50%, and that more accurate EF's can be estimated at 49% for SS and 13% for SI. It is recommended that future NH_3 emission measurements are complemented with N recovery measurements to realize more accurate estimations of emissions.

Abbreviations: ANR, apparent nitrogen recovery; EF, emission factor; SI, shallow injection; SS, surface-spreading; TAN, total ammoniacal nitrogen; TS, trailing-shoe.

Introduction

Ammonia (NH_3) emission after manure application to farmland contributes significantly to anthropogenic NH_3 emission and its potentially harmful effects on the environment through eutrophication and acidification (see introduction in Sintermann et al., 2012). In response, increasingly strict environmental policies are developed internationally to reduce this emission. In a few countries (Denmark, the Netherlands), this has even resulted in a ban on surface-spreading (SS) of liquid manure and the mandatory use of low-emission manure application methods, such as shallow injection (SI). Other European countries may follow this approach, in order to comply with national emission ceilings set by the European Commission (EC, 2001; Potočnik, 2013). In case of the Netherlands, manure application by SI is mandatory on most soil types. Even with SI, roughly half of the calculated total NH_3 emission from nitrogen (N) excreted by confined dairy cattle is emitted after manure application to grassland; the other half is emitted from the barn (Velthof et al., 2009). Since the introduction of mandatory SI in the Netherlands around 1995, there has been controversy about the effectiveness of this method to reduce NH_3 emission and about the accuracy of the official

manure NH_3 emission factors (EF's) that are used. At present, there are still dairy farmers who refuse to apply manure by SI and apply by SS instead, even to the point of prosecution. Recently, the controversy was revived following the release of preliminary results from a small field experiment by Nienhuis and Lantinga (2012). From these results, it could be concluded that the maximum EF after SS was 7% of total ammoniacal N (TAN) applied with liquid cattle manure on grassland, only one-tenth of the official Dutch EF (74%). Although a scientific report was unavailable at the time, the preliminary results and conclusions have led to an intense public debate. The official Dutch EF's for both SS (74%) and SI (19%) (Velthof et al., 2009) are questioned and Dutch parliament has called for scientific review and investigation into this matter (Dijksma, 2013). The preliminary results of Nienhuis & Lantinga (2012) are not the only results to initiate discussion. Internationally, also doubt has risen about the accuracy of measured NH_3 emissions after manure application to farmland. Based on extensive analysis of international emission measurement datasets, Sintermann et al. (2012) suggested that current NH_3 emissions after manure application may be overestimated, and called for new measurement series in order to validate various measurement approaches against each other and to derive revised inputs for inclusion into emission inventories. However, a comparison of various measurement approaches alone will not be sufficient, because such a comparison is relative by nature and not suitable for validation. An alternative approach, with the use of other data than emission measurements, may be necessary to validate the accuracy of measured emission levels. In this paper, such an alternative approach is presented and discussed.

Methods

Emissions can be estimated more accurately when differences in emission between different manure application methods are compared with differences in N recovery from applied manure by crops. Because N not volatilized as NH_3 is largely available for crop uptake, the results of this comparison indicate the level of misestimation (if present) and can also be used for more accurate estimation. This approach is demonstrated for the case of application of liquid cattle manure by SS or SI on grassland grown in a temperate climate. This case was chosen because the most relevant data available are from field experiments with NH_3 emission or N uptake measurements carried out under these conditions. In addition, the two countries (Denmark, the Netherlands) with internationally the strictest environmental policies regarding manure application (e.g. a ban on SS) are located in a temperate climate zone. The manure application methods referred to in the present study are described in detail by Huijsmans et al. (2001). The first step in the proposed approach is to make an inventory of measured differences in apparent N recovery (ANR) between SS and SI. ANR is defined as the difference in N uptake between a crop with manure application and the same crop without manure application, expressed as a percentage of total manure N applied. EF is defined as the percentage of TAN (NH_4-N) in applied manure that volatilizes as NH_3. In liquid cattle manure, roughly half of the N is present in mineral form (TAN) and the other half in organic form (Sörensen, 2004). TAN is largely directly available for e.g. plant uptake and NH_3 emission; organic N slowly mineralizes into TAN over time (Sörensen, 2004).When the difference in ANR between manure application methods is known, a corresponding difference in EF can be derived, using the percentage of TAN in applied total manure N. When, for example, the difference in ANR is 10 percentage points (pp.) and TAN is 50% of total manure N, the derived difference in EF is 20 pp. (= 10 pp. / 0.5). However, TAN applied with manure is not only accounted for by crop N uptake and NH_3 emission. Mineral N, such as TAN, can also be temporarily immobilized in the soil (Sörensen, 2004; Burger and Venterea, 2008), lost by leaching to the ground and surface water (Barraclough et al., 1983; Schröder et al., 2010) or lost in gaseous form other than NH_3 (Egginton and Smith, 1986; Velthof et al.,

1997). When manure application methods differ in the percentages of applied N accounted for by these pathways, the observed differences in ANR, and the derived differences in EF, may be a misestimation of the differences in actual EF's. Therefore, before EF's are derived, an important step is to first discuss potential differences between application methods in N immobilisation, N leaching, and N volatilisation other than NH_3, and correct the difference in ANR, if necessary. Following steps are to use the difference in ANR to derive a difference in EF, compare this derived difference in EF with the measured difference in EF, and determine the level of misestimation in measured EF's (if present). For example, a difference in derived EF's that is 30% smaller than the difference in measured EF's suggests that actual EF's are only 70% of measured EF's. A final step is to use the percentage of misestimation to estimate more accurate EF's for both SS and SI. An important premise for this approach to be accurate is that the level of a misestimation of emission does not vary between manure application methods. Emission data used in the present study were measured with the IHF method (Huijsmans et al., 2001; Sintermann et al., 2012). This approach is internationally considered to be very robust, since it is independent of surface characteristics and the state of atmospheric diffusion (Sintermann et al., 2012). The premise therefore has credibility.

Results and discussion

Differences in apparent nitrogen recovery between application methods

In a five-year field experiment on two different sites (sandy soil) in the Netherlands, first-year ANR of grassland was on average 26% after SS and 44% after SI of regular liquid manure, a difference of 19 percentage points (Groot et al., 2007). Groot et al. (2007) applied on average about 150 kg total N ha^{-1} annually, split in three different applications. Because the manure was applied on the same plots during the five-year period, calculated ANR's also include possible residual effects in later years. In a four-year field experiment on one site (sandy soil) in the Netherlands, first-year ANR was on average 24% for SS and 37% for SI, a difference of 13 pp. (Schröder et al., 2007a). This difference increased to a maximum of 15 pp. in the second year and later (Schröder et al., 2007a). Schröder et al. (2007a) applied on average about 300 kg total N ha^{-1} annually, also split in three different applications. In a three-year field experiment on three different sites in Ireland (sandy loam, loam, clay soil), first-year ANR from spring- and summer-applied manure was on average 22% for SS and 29% for TS (application by the trailing-shoe method), a difference of 7 pp. (Lalor et al., 2011). The difference in ANR between SS and TS is smaller than between SS and SI, e.g. because the contact area between manure and the ambient air is larger for TS compared to SI (Huijsmans et al., 2001). This results in higher NH_3 emission for TS compared to SI (Huijsmans et al., 2001) and consequently a lower ANR. When average differences in measured EF's between SS and TS, and between SS and SI, are known, the difference in ANR between SS and TS can be corrected to reflect the difference between SS and SI. The most extensive dataset of original emission measurements after liquid (cattle) manure application on grassland (see inventory by Sintermann et al., 2012) is provided by Huijsmans et al. (2001). Using these data, average EF's can be calculated at 69% for SS, 27% for TS and 11% for SI. The difference in EF between SS and SI is therefore estimated to be 38% higher than the difference in EF between SS and TS. Since only about 50% of total manure N consists of TAN, the difference in ANR should not be corrected by 38%, but by half this amount (19%).When the earlier found difference in first-year ANR between SS and TS of 7 pp. is corrected for this 19%, the corrected difference between SS and SI is estimated at 8 pp., considerably lower than the 13 pp. reported by Schröder et al. (2007a). Lalor et al. (2011) applied on average 105 kg total N ha^{-1} annually (in spring or summer), without splitting. In a three-year field experiment on one site (clay soil) in Canada (British Columbia Coast), first-

year ANR from spring- and summer-applied manure (early application) was on average 17% for SS and 29% for TS (Bittman et al., 1999). After correction for the earlier mentioned 19%, the difference in first-year ANR was about 14 pp., in line with the 13 pp. found by Schröder et al. (2007a). Bittman et al. (1999) also applied manure in autumn. These results are not discussed here, because the remaining growth period was considered too short for sufficient measurement of residual ANR. Bittman et al. (1999) applied on average about 100 kg total N ha^{-1} (low rate) or 200 kg total N ha^{-1} (high rate) annually (in spring or summer), without splitting. Other studies with ANR measurements after manure application by SS and SI seem to have limited use within the scope of the present analysis. In two studies carried out in Finland, the growing season was shorter, colder and dryer compared to temperate climate conditions, and only part of it was used for ANR measurements (Mattila et al., 2003) or ANR data were not provided (Uusi-Kamppa and Mattila, 2010). Mattila et al. (2003) also applied unusually high manure rates, up to 62 Mg ha^{-1}, applied at once. In an older study from the UK, the applied manure had an unusually low dry matter and TAN concentration, possibly the cause of the observed atypical decrease in ANR of SI compared to SS (Misselbrook et al., 1996).

Differences in nitrogen immobilization between application methods

Manure TAN immobilized in the soil can mineralize in the course of time and become available for plant uptake (Sörensen, 2004; Burger and Venterea, 2008). In two studies found, the difference in ANR between SS and SI increased over time, both within the growing season (Schils and Kok, 2003) as well as over growing seasons (Schröder et al., 2007a). This observation is explained by a higher initial immobilisation of applied TAN after SI compared to SS. Over time, the extra immobilised TAN re-mineralises, and the difference in ANR between SS and SI increases. A focus on the difference in first-year ANR can thus result in a misestimation of potential differences in ANR and derived differences in EF between application methods. When manure is applied on the same plot for several consecutive years (Groot et al., 2007), differences in calculated annual ANR's also include residual differences in later years. Only the results from studies with ANR measurements for at least two consecutive years fully represent the potential differences in ANR due to differences in NH_3 emission (see results of Schröder et al., 2007a). These potential differences vary between 15 and 19 pp. (Groot et al., 2007; Schröder et al., 2007a).

Differences in nitrogen leaching between application methods

When more TAN volatilises as NH_3, less TAN is available for leaching to the ground and surface water. If SS and SI differ substantially in leached TAN fraction, the difference in EF, as derived from differences in ANR can be misestimated. However, no indications for meaningful differences were found in the experiments previously discussed. Particularly in the experiment with very low TAN application rates (Groot et al., 2007) (20 to 30 kg TAN ha^{-1} per application), this effect is likely to have been small to negligible. In a two-year field experiment with the use of $^{15}NH_4$-N as a tracer, N loss by other pathways than NH_3 volatilization was not observed after application of 35 to 59 kg TAN ha^{-1} (Hoekstra et al., 2010). In the three-year field experiment by Bittman et al. (1999), the average difference in first-year ANR between SS and TS was not affected by a doubling of the rate of spring- and summer-applied manure. If there had been meaningful N leaching, ANR would have decreased for both SS and TS. In a three-year field experiment with much higher application of mineral N (250 kg N ha^{-1}) on permanent grassland than in the previously discussed studies, leached N was only 1.5% of applied N (Barraclough et al., 1983). In that study, N was applied by mineral fertilizer NH_4NO_3. The risk of N leaching is generally much larger for NO_3 than for NH_4 (= TAN) (Brown et al., 1982; Mancino and Troll, 1990). Regular application of

organic N with manure can increase the risk of N leaching over time, even at relatively low application rates (De Boer et al., 2012). However, the rate of applied organic N is the same for SS and SI and it is unlikely that the decomposition of organic N differs between SS and SI. Therefore, the rate of N leaching from applied organic N will be comparable and will not affect differences in ANR or derived differences in EF between SS and SI.

Differences in nitrogen volatilisation (other than NH_3) between application methods

When more manure TAN volatilises as NH_3, less TAN is available for gaseous N emissions other than NH_3. When SS and SI differ substantially in the fraction of TAN lost in gaseous form other than NH_3, the difference in EF as derived from differences in ANR can be misestimated. However, no indications were found in literature for meaningful differences between SS and SI in the fraction of TAN lost in gaseous form other than NH_3. The N_2O emission from liquid manure applied to permanent grassland is usually very low, with on average 0.1% of total manure N lost after SS and 0.3% after SI or TS (Velthof and Mosquera, 2011). Relevant studies with measurement of gaseous N emissions other than NH_3 or N_2O are scarcely available. In one study found, total gaseous N losses due to denitrification (which includes N_2O, N_2 and NO_x) were 0.2% or 0% after application of about 90 kg total manure N with SS or SI, respectively (Velthof et al., 1997). These differences were not significant. In another study, net gaseous denitrification losses, following manure applications of 100 to 200 kg total N ha^{-1} by SS, were on average absent (Egginton and Smith, 1986). Klein et al. (1996) recorded net denitrification losses of 3.5% of injected total manure N, but only after deep injection (15 cm) of a very large amount of liquid cattle manure (365 kg N ha^{-1}), at once, during wintertime (December). In the Netherlands, grassland does hardly grow or absorb TAN during winter, and high soil moisture contents during that time greatly enhance denitrification and the associated gaseous N losses. The results of Klein et al. (1996) are therefore not relevant for the present study.

Validation and correction of measured NH_3 emission factors

It can be concluded that manure application methods SS and SI do not substantially differ in the percentages of applied TAN accounted for by other pathways than crop N uptake and NH_3 emission, except for N immobilisation. This effect is however accounted for by using the longer-term difference in ANR between SS and SI. Therefore, correction of the earlier reported differences in ANR between SS and SI is not necessary. Weighed averages for the differences in ANR between SS and SI can be calculated using the number of combinations per study of: experimental years x application moments within year x sites. The weighed average of the difference in ANR is then calculated at 11 pp. (8 to 14 pp.) for the first year of manure application and 18 pp. (15 to 19 pp.) when residual effects are included. Using the TAN concentrations in total manure N, as reported in the discussed studies, the weighed averaged differences in ANR translate into a difference in derived EF of on average 22 pp. (16 to 28 pp.) in the first year of manure application and 37 pp. (31 to 39 pp.) when residual effects are included. The latter difference also represents the average lower limit of NH_3 emission after application of liquid dairy cattle manure by SS on grassland, because EF after SI is at least 0%. The difference between the average measured EF for SS and SI in the previously discussed emission dataset (Huijsmans et al., 2001) is 58 pp. Thus, the difference in derived EF's is in this case only 63% of the difference in measured EF's. This suggests that these measured EF's are overestimated and that more accurate EF's can be estimated at on average 43% for SS and 7% for SI. In case of the Netherlands, the difference between official (measured) EF's for SS (74%) and SI (19%) is 55 pp. (Velthof et al., 2009), 50% higher than the difference in EF's derived from ANR data. Thus, more accurate EF's can be estimated at 49% for SS and 13% for SI.

Differences in experimental conditions between discussed studies

The use of data from different studies, carried out under different experimental conditions, may have affected the results of the comparison of measured and derived EF's. Important aspects to consider are characteristics of the applied liquid manure and manure application methods. Manure characteristics that can influence EF are dry matter (DM) and TAN concentration (Huijsmans et al., 2001). In the study used for data on measured EF's (Huijsmans et al., 2001), average DM and TAN concentration were 77 and 2.15 g kg^{-1} of fresh manure, respectively. Manure DM concentration in the studies used for ANR data ranged between 61 and 77 g kg^{-1} of fresh manure and TAN concentration between 1.27 and 1.85 g kg^{-1} of fresh manure. The composition of manure used in the study by Schröder et al. (2007a) is reported in Schröder et al. (2007b). The two most important studies used for ANR data were Groot et al. (2007) and Schröder et al. (2007a). Average DM concentration (77 g kg^{-1}) in manure applied by Schröder et al. (2007a) was similar to DM concentration in manure applied by Huijsmans et al. (2001), but TAN concentration (1.85 g kg^{-1}) was lower. DM (66 g kg^{-1}) and TAN (1.73 g kg^{-1}) concentrations in manure applied by Groot et al. (2007) were both lower compared to Huijsmans et al. (2001). It is possible that these lower concentrations decreased the EF after SS and the difference in EF's derived from these studies compared to the measured difference in EF's by Huijsmans et al. (2001). This would mean that the suggested overestimation may be smaller than observed. On the other hand, other factors than manure characteristics DM and TAN may have had a similar or larger opposite effect on the difference in derived EF's. For instance, in the study by Groot et al. (2007), the derived difference in EF between SS and SI was 23 pp. on one sandy soil and 56 pp. on the other. The distance between these sites was only 1 km (Groot et al., 2007). Differences between studies in characteristics of manure application methods may also have affected the difference between measured and derived EF's. It has been suggested that in the Netherlands the EF for SI has increased over time, due to a decrease in depth of the injection slots (Huijsmans and Schils, 2009). This may have resulted in a decrease over time of the difference in measured EF between SS and SI. Because the studies used for ANR data are of more recent date than the study used for emission measurement data (Huijsmans et al., 2001), part of the observed overestimation may have been caused by this effect. However, in case of the Netherlands, the official EF for SI has already been corrected for this possibility (Huijsmans and Schils, 2009). Nevertheless, the difference between these measured EF's is still 50% higher than the difference between EF's derived from ANR data. The possible effects of differences in experimental conditions between the different studies give reason for some caution regarding the reported level of overestimation and re-estimated EF's, but, more importantly, underscore the importance of combined measurement of emissions and ANR's in future experiments.

Conclusion

A comparison of differences in measured EF's and EF's derived from measured ANR data is useful to realize a more accurate estimation of actual NH_3 emissions after manure application to grassland. The results of the present study suggest that EF's measured by the IHF-method are indeed overestimated, as previously suggested by Sintermann et al. (2012). However, this overestimation is not as large as suggested by the (preliminary) data of Nienhuis & Lantinga (2012), and the lower limit of NH_3 emission after manure application by SS on grassland seems firmly set at on average 37% of manure TAN. Internationally, application of the approach proposed in the present study may give cause for a re-estimation of EF's and adjustment of national emission inventories and environmental policies. In future field research, a comparison of EF's derived from differences in ANR, and EF's measured with different methods, may be useful to determine which emission measurement method is most

accurate. It is recommended that future NH_3 emission measurements after manure application are complemented with ANR measurements to realize more accurate estimations of these emissions.

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