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ORIGINAL RESEARCH

Comparison of ammonia emissions from animal wastes and chemical fertilizers after application in the soil

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Abstract

Background Application of different chemical fertilizers and manures is a major source of ammonia (NH_3) emission. The rate and total amount of NH_3 emission are related to different parameters such as climatic conditions, soil characteristics and kind of fertilizer. The current study has indicated the NH_3 emission from bovine slurry, pig slurry and ammonium nitrate fertilizer after application on two soils. Two different methods were used to measure NH_3 emissions: the method that use acid traps and the method that use photoacoustic infrared gas analyzer.

Results In both soils the rate of NH_3 emission was the greatest from the denser bovine slurry, declined in the pig slurry followed by the ammonium nitrate treatment and the control. The rate of soil infiltration could be the main factor that explained these differences. For all treatments the amount of total NH_3 losses reduced in the more acidic soil. For all fertilizers the highest NH_3 fluxes were measured in the first hours after spreading. A good agreement observed between the two methods is used for determining of NH_3 emission. The use of a multi-gas monitor (MGM) is simple and accurate and produces a continuous series of NH_3 concentration in time.

Conclusion The rate and amount of NH_3 emission was related to the kind of fertilizers and interaction of these treatments with soils. The results of current study confirmed that comparison of chemical fertilizers and slurry for NH_3 emission is difficult because the reaction of these two groups of fertilizer is totally different.

Keywords Ammonia emission · Laboratory methods · Mineral fertilizer · Slurry

Introduction

Ammonia (NH_3) may be released into the atmosphere from basically all ammonium-containing products. Livestock and especially animal manures are the most important sources of NH_3 emissions in Europe, followed by the application of mineral nitrogen fertilizers (Leip et al. 2011). The increase in animal stocking and in the price of chemical fertilizers encourage farmers to use animal manure and slurry as an option to reduce the use of commercial fertilizers. However, the handling and spreading of these fertilizers may pose an agronomic and environmental risk, not only because of leakage of nitrate to ground waters but also because of gaseous losses of NH_3 (Asman 1992). Ammonia can form secondary particulate matter in the

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atmosphere that may have adverse effects on human health (Moldanová et al. 2011).

Ammonia emissions from manure applied to the soil are produced primarily by physical and chemical processes and secondarily by biological ones (Monaco et al. 2012). Ammonia losses from manure are harmful from the agronomic point of view, because they decrease the amount of manure N available for the crop (Smil 1999). Olivier et al. (1998) estimated that about 70 % of global NH_3 emission is related to food production and, in particular, to manure management.

A comprehensive understanding of the post application fate of fertilizers is essential for the development of best management practices that aim to minimize off-site transport and maximize nutrient use efficiency. Different variables affect both the rate and extent of emissions following soil application of manures (Meisinger et al. 2001). The dominant factors influencing losses can be categorized as: manure characteristics (dry matter content, pH, NH_4–N content), application methods (incorporation, time of application), soil properties (soil moisture, soil texture, soil pH), and environmental factors (temperature, wind speed, rainfall, relative humidity).

A number of techniques have been developed to quantify NH_3 emission. Accuracy and mechanism of these methods are very different. Such techniques fall generally in two groups: micro-meteorological methods (usually used for large scale areas) and enclosure methods (commonly used on small plots for comparative experiments). Chamber methods that belong to second category are usually used for measuring emission at the small scale both in the field and in the laboratory. Three measurement schemes are commonly used for the chamber methods: the open chamber, the closed static chamber and the closed dynamic chamber. All methods employ an inverted chamber covering a small area of soil. The lower edge of the chamber usually is inserted into the soil to a shallow depth.

The analysis of NH_3 emission have been carried out using different methods as acid traps or direct measurement through a multi-gas monitor (MGM). Between them, using MGM is easier because this system provides a real-time analysis of NH_3 concentration (Dinuccio et al. 2008). Experiments for measuring NH_3 emissions are usually carried out in the field where soil and moisture conditions and other environmental factors are variable and hard to control. Unfortunately, little effort has been made to standardize the laboratory methods for NH_3 emission measurement. The objectives of this experiment were to measure NH_3 emissions from different slurries using closed dynamic chambers with both acid traps and MGM method under controlled environmental conditions and assessing the influence of two different soils on modifying NH_3 emissions.

Materials and methods

The amount of volatilized NH_3 after surface application of different fertilizers in two soil types was assessed under constant and controlled environmental conditions using dynamic chamber technique (Roelle and Aneja 2002). The soils used in the experiment presented different physical and chemical characteristic (Table 1) and were representatives of arable soil types of the western Po river plain (Northern Italy). In particular, Poirino soil came from a farm field cultivated mainly with maize for grain in rotation with wheat receiving chemical fertilizations, while Tetto Frati soil (TF) was collected from an experimental field continuously cultivated with maize for grain fertilized with cattle slurry and urea. The soil samples were collected from the tilled top 20 cm layer on, air-dried and sieved using 2 mm. Cylindrical glass jars (3.1 l) were filled with 1.1 l of each soil type moistened to reach the field capacity (FC). In particular, 1595 and 1475 g of dry soil was moistened with 367 and 428 ml of deionized water for Poirino and TF soil, respectively. Soil moisture content at FC (−33 kPa) was measured on 4 replicates for each soil type using pressure plates and was equal to 0.23 and 0.29 g of H_2O per g of dry soil for Poirino and TF, respectively. Bulk density of soils at FC were measured by drying 100 cm^3 of soil (four replicates) at 105 °C for 3 days, and were equal to 1.45 and 1.34 g of dry soil per cm^3.

Organic fertilizers used in the experiment were pig slurry (PS) and bovine slurry (BS). Slurries were collected from storage facilities of two farms few days before the beginning of experiment, stored at 4 °C and analyzed for the main characteristics (Table 2). Fertilizers were applied at a rate of 85 kg ha^{-1} of total N. In particular, 34.8 and 67.4 g of PS and BS were gently distributed on the surface of the jars (154 cm^2), providing 131 mg jar^{-1} of total N and 95 and 65 mg jar^{-1} of ammoniacal N for PS and BS, respectively. For each soil type, a treatment fertilized with ammonium nitrate at the rate of 85 kg ha^{-1} of total N was also prepared. Ammonium nitrate was solubilised in 15 ml deionized water and distributed on the soil surface. In addition, one extra jar in which BS was

Table 1 Physical and chemical properties of soils

Soil	Sand (%)	Silt (%)	Clay (%)	CEC Meq/100 g	pH	OM (%)	N (%)	C/N ratio
Poirino	15.8	75.5	8.7	16.7	6.2	2.4	0.14	9.6
TF	48.4	43.1	8.5	12.4	7.9	2	0.17	7

Table 2 Physical and chemical properties of the slurries

Slurry type	Dry matter (%)	pH	N (%)	TAN (%)
Bovine	5.8	8.5	19	10
Pig	2.3	8.0	38	27

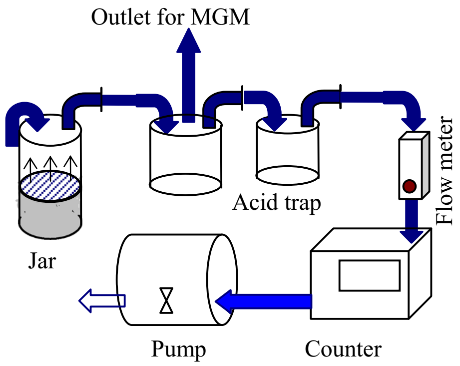

Fig. 1 Schematic diagram of the measuring system

incorporated in Poirino soil was also analyzed. For each soil type, an unfertilized soil was used as control treatment. One replication for each treatment was measured in three different sessions of measurement in a growth chamber properly equipped for NH_3 emission assessment using open dynamic chamber method. Each session was conducted at 25 °C under controlled environmental conditions. Ammonia concentration in the incoming air (background) was always measured and found negligible (0.08 ppm).

Immediately after fertilization, jars were closed with air-tight lids prepared with one input and one output port directing air into the headspace and connecting the jars to the measurement system (Fig. 1). In particular, each jar was connected first to an expansion bottle and then connected in an air-tight way with a Drechsel bottle, flow meter, gas meter and pump. The flow rate ($2\ l\ min^{-1}$) was chosen to ensure a minimal exchange rate, but also to prevent rapid drying of the soil surface. It corresponded to an air renewal rate of one headspace volumes min^{-1} with an average air flow rate of $0.12\ m^3\ h^{-1}$. The air flow rate was monitored over each jar using one volumetric air flow meter per jar. From the expansion bottles another outlet tube was inserted for measuring of NH_3 concentration using photoacoustic infrared gas analyzer (Innova 1412).

In the acid trap method, the air stream passing through the jar and the expansion bottle arrived to a NH_3 scrubber containing 100 ml of 0.1N H_2SO_4, in which absorbed NH_3 was converted to ammonium sulfate. All of the acid samples were stored at 4 °C until the analysis. The amount of NH_3 absorbed in each scrubber was determined using an ionometer (Ion lab, WTW). The NH_3–N concentration in the outgoing air was calculated from the data of ionometer multiplied by the volume of acid trap.

Measurements with acid traps were carried out in four intervals of 5 h during the first 4 days after slurry application. Measuring for 4 days was considered enough to reveal the possible differences in NH_3 emission, because according to earlier studies NH_3 emission is at its highest rate on the day of application and declines sharply in the following few days (Yang et al. 2003). The total amount of emitted NH_3 during the 4 days was derived by interpolation of the integral form of a power curve of the amount of NH_3 trapped in the Drechsel bottle during the four measurements.

Simultaneously, measurements with MGM were carried out during the first 2 days after fertilization, when the NH_3 emissions were expected to be high for fertilized treatments. Gas sample suction for MGM started approximately 30 min after the closing of chamber to allow stabilization of NH_3 concentration in the expansion bottle and was repeated three times for treatment during each interval of measurement. Before each measurement, Teflon tubes connecting the system to MGM were cleaned with background air flush.

The NH_3 readings by MGM (mg/l) were multiplied by a correction factor for the atmospheric pressure. NH_3 concentrations (mg/l) were converted into the flux of NH_3 leaving each jar as follows:

$$\mathrm{FNg} = \text{air volume} \times |\text{conc}| \times 10^{-6} \times \rho\mathrm{NH}_3 \times (14/17)$$

where FNg is the NH_3–N flux in mg N per jar; air volume is the throughput of air during one measurement; |conc| is the value of the corrected volume concentration (vol. mg/l) of NH_3; ρNH_3 is the density of NH_3 in $mg\ l^{-1}$ ($\rho NH_3 = 696\ mg\ l^{-1}$; 25 °C, 1013 hPa).

The total amount of NH_3 emitted during each interval of measurement was calculated separately for each treatment by the integration on time basis of the measurements carried out with MGM, using linear, exponential and power function. The results were then compared with total amount of NH_3 emitted as measured with acid traps. Calculations and drawing the trend lines was done with using Microsoft Excel-2003, and SPSS.

Results

Comparison of manures using the acid trap method

Immediately after the application of fertilizers, NH_3 emission started and continued up to the end of the experiment. Based on Kruskal–Wallis test ($P \leq 0.05$) differences among treatments were significant. Until the third day of experiment, the ranking of treatments did not change and the rate of NH_3 emissions were highest in BS, followed by PS and ammonium nitrate, and were very low in control for both soils. Incorporation of BS strongly reduced emission to the level of control treatment. On day 4 the ranking of the treatments was different, but emissions were very low (Fig. 2).

One day after application of fertilizers, NH_3 fluxes declined sharply by about sevenfold in slurry treatments and threefold in mineral fertilizer (Fig. 2). Ammonia fluxes had fallen to the lowest levels by the third or fourth day depending on the treatments. Based on results the most NH_3 losses occurred during the first day after application of fertilizer.

With respect to their NH_3 emissions both the slurry treatments showed systematic time course patterns over the 4 days. The soils fertilized with pure mineral fertilizer, also showed similar patterns, but at much lower levels. The emission rate did not change in time from the manure incorporated treatment and the control treatment, that showed a low and steady flux during the whole experiment.

The total emissions of NH_3 during the experimental period were calculated using integral of measured emission rate with acid trap method (Fig. 3a) and reported as a percentage of ammoniacal nitrogen applied (Fig. 3b). In both the cases Kruskal–Wallis test reported significant differences among treatments.

The rate and total amount of NH_3 emission was related to kind of fertilizers. In this experiment the range of NH_3 emission from BS was 39–82 % of total ammoniacal nitrogen (TAN) applied and the amount of emitted NH_3 in PS and ammonium nitrate varied from 13–38 to 2–18 % of TAN, respectively. This shows that the higher dry matter content of the slurry the higher emission rate.

Figure 3 shows that there was a similar ranking of treatments for total amount of emitted NH_3 and of the ratio between emitted NH_3 and TAN. In other words, in our experiment the amount of TAN in the different fertilizers did not affect the final NH_3 emissions. The amount of total

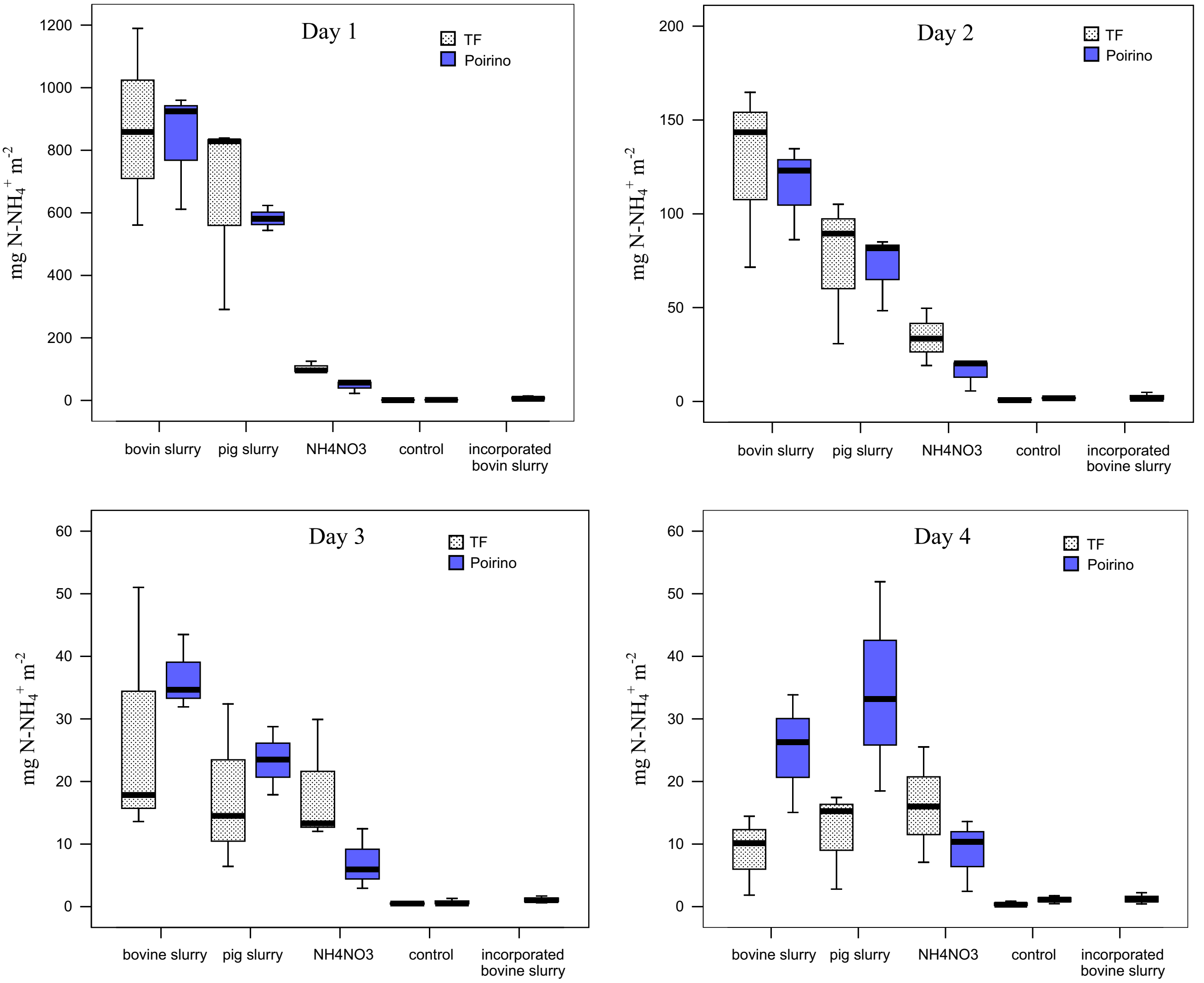

Fig. 2 Ammonia emission from different treatments during measurement times in each day (measured with acid traps)

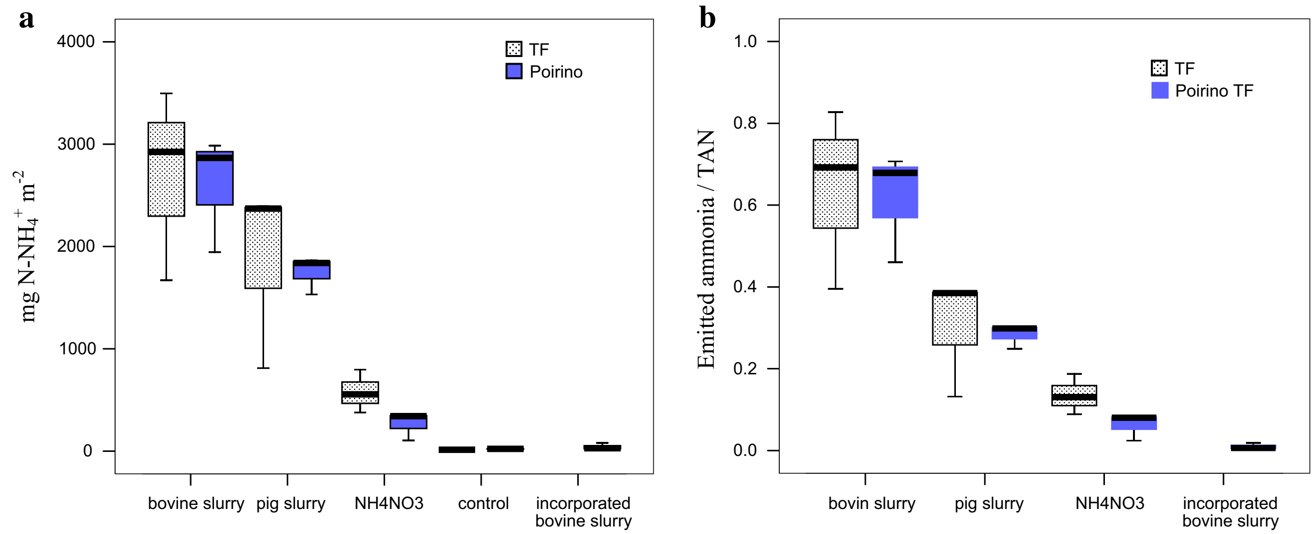

Fig. 3 Total amount of emitted ammonia from different treatments during the experiment (**a**), and ratio between emitted ammonia and total ammoniacal nitrogen (TAN) in different treatments (**b**) (measured with acid traps)

NH_3 losses in the calcareous TF soil was higher than in the more acidic Poirino soil for PS and ammonium nitrate treatment, the two fertilizers that are expected to better infiltrate into soil because of lower dry matter. Instead no differences observed for the BS, which was the fertilizer characterized by higher dry matter.

Comparison of the two methods for ammonia measurements

Using MGM the highest NH_3 fluxes were measured during the first hour following the application of fertilizer in all the fertilized treatments (Fig. 4). But it is likely that the fluxes immediately after application were still greater, due to the delay (about 30 min) in the beginning of measurements. The time variation patterns of NH_3 concentration in the air fluxes were similar in both the methods. In the second day after application of fertilizers, NH_3 fluxes declined sharply by about sixfold in slurry treatments and less than twofold in mineral fertilizer (Fig. 4).

For the interpolation of the amount of emitted NH_3 measured with MGM in each interval, three different mathematical models were used. A strong linear relationship was observed between the NH_3 concentration by acid trap and MGM methods (Fig. 5), but in general, the amount of emitted NH_3 measured with MGM was higher than that measured by acid trap. Comparison of the three methods used for calculation of the total amount of emission showed that they are all acceptable, but the best method for representing data of MGM in short times (5 h) is using the

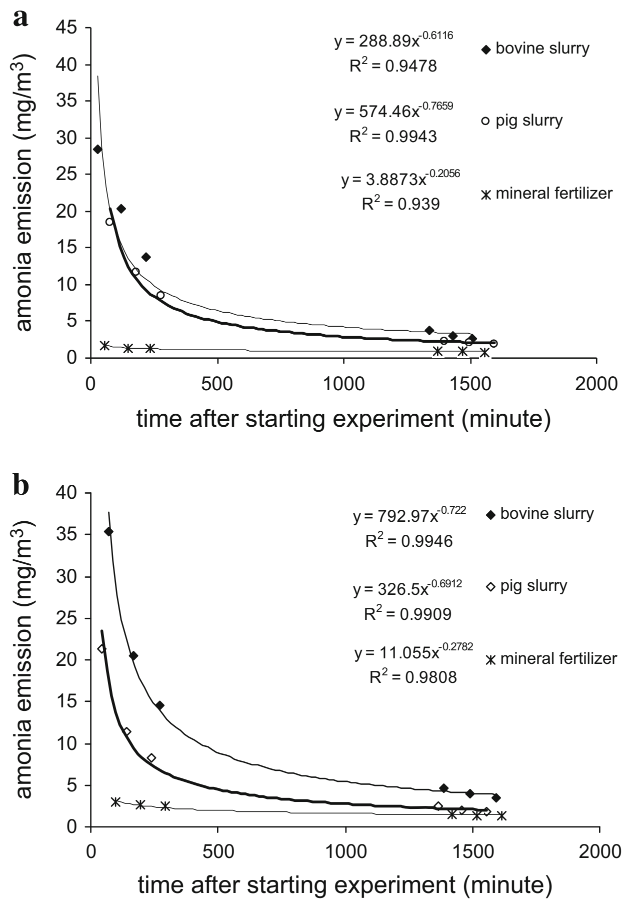

Fig. 4 Rate of ammonia emission measured with MGM in **a** Poirino and **b** TF soil

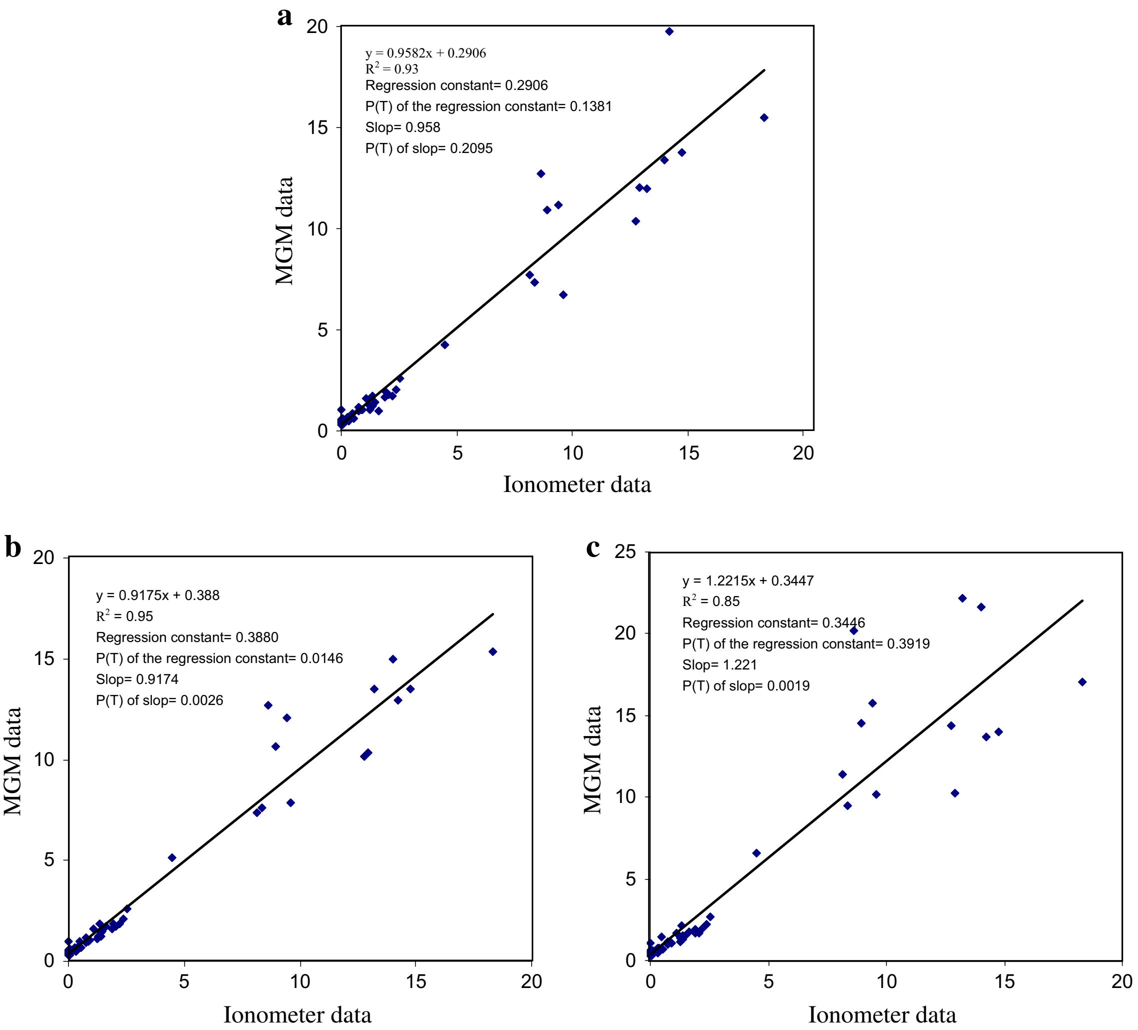

Fig. 5 Regression between amounts of ammonia emission measured by ionometer and derived from MGM measurements interpolated with **a** linear, **b** exponential and **c** power function

linear function, whereas the use of a linear function for representing the trend of NH_3 emission in longer duration (day) is not justified.

Discussion

The highest rate of NH_3 fluxes was measured using MGM in the first day and in the second day after application of fertilizers NH_3 fluxes declined sharply. Huijsmans et al. (2001) reported that the rate of NH_3 emission decreased with time. NH_3 emission also decreases with a reduction in slurry water content because the water content initially decreases due to infiltration of the slurry liquid into the soil.

Meisinger et al. (2001) reported that losses of NH_3 through emission are very rapid during the first 6–12 h after application. Results of previous study showed that 30–70 % of the total NH_3 loss from cattle slurry occurred in the first 4–6 h, and 50–90 % in the first day (Stevens and Laughlin 1997); however, this result was related to the application of slurries in grassland and is not totally comparable with our results obtained under controlled conditions.

The TAN concentration of pig slurry was higher and the dry matter content was lower than that of the bovine slurry, as it is often the case (Dinuccio et al. 2008), but in this experiment the higher amount of NH_3 emission was observed when BS was applied. This result is probably related to interaction between soil and slurry.

Higher dry matter content of the bovine slurry decreased infiltration into the soil, and it increased the chances of NH_3 losses, as it was also indicated by Stevens et al. (1992). The slurry dry matter content has been shown to be an important factor in determining the NH_3 emission potential. Sommer and Olesen (2000) showed a linear relationship between cattle slurry dry matter content and NH_3 emission in the range 4–12 % DM, but outside this range dry matter had a little effect on NH_3 emission. Smith et al. (2000) reported a similar linear relationship but in a shorter range (2–5 % of slurry dry matter) and concluded that for every 1 % increase in slurry dry matter, NH_3 losses increased about 6 %. In the MANNER model that was developed for predicting NH_3 emission from manure, it was suggested that for every 1 % increase in manure DM, NH_3 loss increases about 5 % (Chambers et al. 1999). In the Poirino soil the difference between total amount of emitted NH_3 from BS and PS was higher than in TF soil (Fig. 2), and this difference probably was related to the soil texture because the importance of slurry dry matter (for NH_3 emission) in soils with a poor infiltration rate is higher (Jarvis and Pain 1990).

Soil pH and cation exchange capacity (CEC) also influenced NH_3 emission. When the fertilizer could infiltrate in the soil, the higher pH of the TF soil enhanced NH_3 emission in comparison with the soil of Poirino. This confirms that lower pH leads to a lower proportion of aqueous NH_3 and therefore decreases NH_3 emission (Li 2000). Huijsmans et al. (2003) and FAO (2001) reported similar results, in which the increase of pH and decrease of CEC could raise NH_3 emission of 10 times. Soil surface pH greatly changes when slurry is applied to soil, due to the different buffering capacities of the soil and slurry. The highest reaction of NH_3 emission to pH was observed when pH ranges from 7 to 10, and NH_3 emission below 7 decreases dramatically (Hartung and Phillips 1994). Using a mechanistic model for estimating the NH_3 emission from slurry after application in soil Genermont and Cellier (1997) pH was the main factor that influenced NH_3 emission. They reported that the reduction of the pH from 7 to 6 resulted in 19 % decrease of NH_3 emission.

The total amount of emitted NH_3 measured in our experiment was close to the values reported in other experiments. Stevense and Laughlin (1997) showed that NH_3 losses from surface applied liquid cattle manure in grassland was in the range 40–70 %, whereas NH_3 losses from poultry litter in pasture ranged from 28 to 46 % of TAN (Marshall et al. 1998).

This research work confirmed the general idea that the amount of emitted NH_3 from mineral fertilizer is less than from slurry. Whitehead and Raistrick (1990) estimated that only 3.4 % of the applied N in chemical fertilizer (five nitrogen compounds) was lost as NH_3. In reporting emissions from fertilizers for the UK, Lee and Dollard (1994) used emission factors of 3 % for ammonium nitrate.

Incorporation of slurry almost suppressed NH_3 emission and in this treatment total amount of emitted NH_3 was negligible. Other researchers also mentioned that incorporation of manure was one of the best methods for decreasing NH_3 emission. Rodhe et al. (2006) reported that by injection of slurry into the soil the emission decreased by 39 % in comparison with the band spreading of slurry. By incorporating and mixing slurries with soil, contact area between the slurry and the air was reduced, and therefore NH_3 emission in comparison with surface application was decreased. Sommer et al. (1991) reported that evaporation of water from the manure lead to an increase of the aqueous NH_3 concentration in the manure and to an increase in NH_3 emission.

Results of this experiment showed that for calculating total amount of emitted NH_3 by MGM, different functions should be used because the trend of NH_3 emission based on the time scale of measurement is different. Regarding the NH_3 emission rate measured with MGM in the first 2 days, power function can represent the data in better comparison to linear functions. These results are in agreement with results of other experiment (Bussink et al. 1996) that showed that the trend of NH_3 emission rate from applied manure was not linear. The results of this experiment are also in contrast with results from Chambers et al. (1997), who observed linear rates of emission from poultry litter for up to 3 weeks after application.

Conclusion

The rate and amount of NH_3 emission were related to the kind of fertilizers and interaction of these treatments with soils. Comparison of chemical fertilizers and slurry for NH_3 emission is difficult because the reaction of these two groups of fertilizer is totally different. The amount of emitted NH_3 from different fertilizers was related to the amount of water added to the soil with the application and consequently to the infiltration of the slurry into the soil. It seems that 4 days are enough for the estimation of the NH_3 emission from slurry, but longer time is needed for chemical fertilizers. The strong linear correlations between the results of the two measurement methods demonstrated that MGM was also utilizable for determination of the total amount of emitted NH_3 in different treatments even though a slight overestimation existed.

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