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ORIGINAL PAPER

Emissions of Nitrous Oxide, Methane and Ammonia After Field Application of Digested and Dewatered Sewage Sludge With or Without Addition of Urea

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Abstract

Purpose By recycling sewage sludge (SS) to productive land, its plant nutrients can be utilised. However, the use of organic fertilisers carries health risks and causes emissions of nitrous oxide (N_2O), methane (CH_4) and ammonia (NH_3). One measure to sanitise SS from human pathogens is addition of NH_3.

Methods Mesophilically digested and dewatered SS treated with urea and stored, or only stored, was applied to arable land in spring and autumn, respectively, and the effects of immediate or delayed incorporation (by 4 h) on emissions of N_2O, CH_4 and, in spring, NH_3 were investigated.

Results N_2O emissions in autumn from soil treated with SS were significantly higher than from soil without SS application (0.09, 1.31 and 0.68 kg N_2O-N ha^{-1} for control, immediate and delayed incorporation, respectively). These emissions were significantly correlated with volumetric water content in soil. Corresponding N_2O emissions in spring were 0.15, 0.57 and 0.41 kg N_2O-N ha^{-1}. Delayed incorporation (0.20 and 0.34 % of added N in spring and autumn, respectively) tended to reduce N_2O emissions compared with immediate incorporation (0.32 and 0.71 % of added N in spring and autumn, respectively). Nitrous oxide emissions from SS were apparently lower after spring than after autumn application, likely because of drier soil and crop uptake of nitrogen in spring. Methane emissions were negative or negligible. Timing of incorporation had no statistically significant effect on NH_3 emissions.

Conclusions Nitrous oxide emissions from soil treated with SS at a rate based on the maximum permissible P level were moderate and CH_4 emissions negligible.

Introduction

Sewage sludge (SS) from municipal wastewater treatment contains nutrients and organic matter that can be beneficial to agriculture. The previous target in Sweden was for at least 60 % of the phosphorus (P) in wastewater to be recycled to productive land by 2015 [1]. In 2012, about 24 % of the SS produced in Sweden was recycled to arable land [2]. The application rate of SS to soil is determined by the P content, the rate should, according to Swedish law, not exceed 110 kg P ha^{-1} during a 5-year period [3]. For chemically precipitated, digested and dewatered SS, this means a low application rate in terms of mass and nitrogen (N). Because of the risk of human pathogens in SS, the Swedish Environmental Protection Agency (EPA) suggests that SS be sanitised before use on land. Two alternatives proposed for such sanitation are thermophilic digestion (50–60 °C) and sanitisation with ammonia (NH_3) [4], which can be achieved by urea (($NH_2)_2CO$) addition. Urea addition also adds to the SS fertiliser value.

Use of organic fertilisers causes emissions of the greenhouse gases (GHG) nitrous oxide (N_2O), and methane

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(CH_4), and also of NH_3, to the atmosphere. The global warming potential for CH_4 and N_2O corresponds to 34 and 298 CO_2-equivalents, respectively, in a 100-year perspective [5]. Sewage sludge contains organic and mineral N, so the risk of N_2O emissions originating from microbial processes in soil fertilised with SS is potentially high. Nitrous oxide can be formed under both aerobically intermediate and anaerobic conditions [6], as a by-product of microbial nitrification and denitrification. The microbes involved in nitrification and denitrification are natural components of the soil microbial ecosystem [7] and the potential activity of both groups is generally high [8]. Methane is produced by methanogens under strictly anaerobic conditions [9]. In soil, waterlogging is usually required for CH_4 formation and hence CH_4 production is mainly limited to wetlands, lake sediments and rice fields. Methane formation in aerated soils is normally low [10, 11]. Emissions of NH_3 involve loss of plant-available N and also contribute to acidification and eutrophication upon atmospheric deposition [12]. Ammonia emissions also comprise fine particulate matter, which is a health issue when inhaled [12]. Depending on the storage method used, organic waste often contains high concentrations of ammonium (NH_4^+), which on application to soil can be volatised to NH_3.

Previous studies on the impact of SS application on emissions of GHG and NH_3 show varying results, ranging from no difference between SS-fertilised soil and unfertilised soil to distinct differences in emissions of one or more of the gases measured [13–21]. However, the production methods for the SS used in those studies varied and included air-drying, grinding, dewatering by centrifuge and digestion. Soil and SS properties, climate, application rate and application method also differed between the studies. In addition, field studies on application of digested and dewatered SS are scarce, especially in climates comparable to those in the present study, since a majority of previous studies have been performed in a laboratory, greenhouse or in a tropical climate.

Many different factors influence N_2O emissions after application of organic fertilisers such as SS, cattle and pig slurry and farmyard manure. Soil moisture [21–23] and precipitation [14, 24] have a major impact on N_2O emissions, with moderate to high rainfall events stimulating N_2O fluxes. Temperature and rainfall generally affect formation and emission of GHG, and both daily and seasonal variations have been reported [14]. The presence of a growing crop can decrease the amount of N_2O emitted [25, 26].

Incorporation within 1 day after application of SS to arable land, e.g. by harrowing, has been proposed as a requirement for SS management in Sweden to reduce both the exposure of humans and animals to SS and to the risk of surface runoff [3]. Incorporation of organic fertiliser into the soil immediately or as soon as possible after application is also a commonly used and effective method for reducing NH_3 emissions [27], and in many regions mandatory by law. However, some claim that incorporation of animal slurry [23, 24, 28] and solid manure [29] into the soil can give higher N_2O emissions than surface application. Other studies have not shown any difference in N_2O or CH_4 emissions from incorporated and surface-applied slurries [30, 31], while yet others report higher emissions of N_2O from solid manure left on the surface than when incorporated [29, 32]. The rate of N applied as organic fertiliser also has an impact on N_2O emissions, as a higher application rate can give higher emissions [23, 33].

It has been shown that ammonia treatment by addition of urea sanitises the SS [34], but the N_2O or CH_4 emissions from such sludge after application to soil has not been published.

The overall aim of this study was to quantify emissions of N_2O, CH_4 and NH_3 from arable land after application of mesophilically digested and dewatered SS. A specific objective was to evaluate the effect on GHG emissions of immediate incorporation of applied SS into soil compared with delayed incorporation. For this, emissions after spring application of mesophilically digested, dewatered, urea-treated and stored SS and after autumn application of mesophilically digested, dewatered and stored SS were measured.

Materials and Methods

Site Description and Experimental Design

The experiments took place in an arable field (59°52′N, 17°31′E) 8.2 km NW of the city of Uppsala, Sweden. Data on air temperature and precipitation during the experimental period were obtained from a weather station (59°50′N, 17°37′E) run by the Department of Earth Sciences at Uppsala University, located about 7 km from the experimental field. During the measurements of NH_3 emissions in the spring experiment, a mobile meteorological station (Adcon 753 and Adcon Combo 730R, Adcon Telemetry, Klosterneuburg, Austria) recorded air temperature 1.5 m above ground, precipitation and wind speed at the site of the experiment. The soil at the site is a silty clay with 10 % sand, 46 % silt and 44 % clay [35], with 3.0 % organic matter content. Other soil properties are presented in Table 1. Winter wheat was grown in the field in the year before the experiment and the field had received only mineral fertiliser during the past 50 years.

In spring and in autumn, field plots were established and SS was applied, after which gas sampling for determination of

Table 1 Soil characteristics before application of sewage sludge (SS) and of mesophilically digested and dewatered SS treated with urea applied in spring and mesophilically digested and dewatered SS without urea applied in autumn

Substrate/time	DM (%)	VS (% of DM)	pH	Tot-N (kg Mg^{-1} DM)	Org-N (kg Mg^{-1} DM)	TAN (kg Mg^{-1} DM)	NO$_3^-$-N + NO$_2^-$-N (kg Mg^{-1} DM)	TOC[a] (kg Mg^{-1} DM)	Tot-P (kg Mg^{-1} DM)
Soil	83.5	5.0	6.9	2.4	2.4	0.006	0.005	19.7	6.2
Spring	26.9	59.0	–	67.1	37.9	29.2	–	319.2	32.5
Autumn	24.6	58.0	–	51.5	36.1	15.3	–	305.7	32.7

DM dry matter, *VS* volatile solids

[a] SS was analysed for Tot-C, but the results are presented as TOC for clarity

N$_2$O and CH$_4$ emissions was conducted. In the spring experiment, NH$_3$ emissions were also measured. Both experiments were organised in one completely randomised block design with three replicates (blocks) per treatment. Different plots were used in the spring and autumn experiments. Each experimental plot measured 6 m × 12 m for the SS treatments and 3 m × 12 m for the unfertilised controls. In spring, SS was applied to the entire experimental plots, while in autumn three areas measuring 1.5 m × 1.5 m were randomly chosen in each experimental plot for SS application. In each experiment, the treatments consisted of application of one type of SS that was either incorporated into the soil immediately after application or incorporated after a 4-h delay. Unfertilised plots were used as controls. In the spring experiment, the digested, dewatered, urea-treated and stored SS was applied on 3 May 2012, while in the autumn experiment the digested, dewatered and stored SS was applied on 8 October 2012. The GHG emissions measurements continued in spring until the crop grew too large for the chambers and in autumn until freezing of the soil. Emissions of N$_2$O and CH$_4$ were measured for 67 days in spring (4 May–9 July 2012) and for 45 days in autumn (9 October–22 November 2012). Emissions of NH$_3$ were measured for 24 h in spring. Soil and air temperature, soil moisture and precipitation were also recorded during the measuring periods.

Soil and Sewage Sludge Sampling and Characterisation

Before SS application in spring, a composite soil sample consisting of 15 subsamples from the entire experimental site was collected at random spots along the diagonal of the field at 0–0.2 m depth for soil characterisation. Soil texture was analysed by the hydrometer method described in Swedish standard 027123-24U2 and dry matter content (DM) and volatile solids (VS) according to [36]. Total nitrogen (Tot-N) was analysed according to SS-ISO 13878 and the N fractions ammonium nitrogen [interpreted as total ammonia nitrogen (TAN)] and nitrate/nitrite nitrogen (NO$_3^-$-N/NO$_2^-$-N) according to SS-ISO 14256-2:2005. Organic nitrogen (Org-N) was calculated as the difference

between Tot-N and the sum of TAN and NO$_3^-$-N/NO$_2^-$-N. Total organic carbon (TOC) was analysed according to SS-ISO 10694 and C/N ratio was calculated as TOC divided by Org-N. Total phosphorus (Tot-P) was analysed according to SS 28311 and pH according to SS-ISO 10390. Additional soil samples for determination of soil bulk density and water content were collected with steel cylinders before SS application in spring and autumn, at two depths (0–0.05 and 0.05–0.10 m) in each plot. During the two measuring periods, soil temperature was recorded at 0.02–0.05 m depth hourly using a temperature sensor with data storage (Tiny Tag, Intab Interface AB, Stenkullen, Sweden). The volumetric water content (vol. WC) of the topsoil was recorded every 10 min with a Theta Probe ML2x sensor (Delta-T Devices Ltd., Cambridge, UK). Both the temperature and moisture sensors were placed in the centre of the experimental area. Volumetric WC is defined in (Eq. 1).

$$\text{vol. WC} = \frac{V_{water}}{V_{total}} \qquad (1)$$

where V_{water} is the water volume and V_{total} represents the volumes of the solid phase and the pore space.

Mesophilically digested and dewatered SS was delivered from Kungsängsverket, the municipal wastewater treatment plant in the city of Uppsala, Sweden (59°50′N, 17°39′E). The SS applied to the soil in spring was sanitised by addition of urea (1.5 wt%) and then stored under a plastic cover for 5 months before application. The SS used in the autumn had been stored under a plastic cover during 13 months before application. Before application, the SS was sampled for characterisation of DM, VS, Tot-N, ammonium nitrogen (interpreted as TAN), Tot-C and Tot-P. The methods for analyses and calculations were the same as those for the soil characterisation except for C/N ratio, which was calculated as Tot-C divided by Tot-N.

Sewage Sludge Application

In the spring, the SS was applied with a JF ST 9500 manure spreader (JF-Stoll, Sønderborg, Denmark) provided with

two horizontal beaters, using a control system for constant application rate developed at the Swedish Institute of Agricultural and Environmental Engineering (JTI) in Uppsala, Sweden. The target application rate was based on the maximum P application rate allowed, i.e. 110 kg P per 5-year period, which corresponded to about 12 Mg SS ha^{-1}. The actual application rate was determined by weighing four trays, each 0.25 m^2, that were placed beside each plot before the spreader passed. The SS was incorporated into the topsoil either immediately or 4 h after application, with a harrow working to a mean depth of 0.04 m. After the delayed incorporation, all plots were sown with spring barley and rolled.

Before the field experiment, the fertiliser spreader was tested and the application rate calibrated with SS from the same wastewater treatment plant as used in the experiment. However, due to quantity restrictions the urea-treated SS could not be tested. During application in the experiment, it was discovered that the urea-treated SS was much stickier than the SS used for calibration. Problems therefore occurred in spring, resulting in lower mean rate than desired and uneven distribution of SS.

In the autumn, the SS was evenly hand-spread on stubble. The target application rate based on maximum allowed P application rate corresponded to 13.7 Mg SS ha^{-1}. The SS was weighed into buckets (one bucket per 1.5 m × 1.5 m plot) before application and incorporated into the topsoil, either immediately or 4 h after application, with a rotary cultivator working to a mean depth of 0.06 m (0.04–0.10 m min–max). The control treatment was cultivated in the same way. In autumn, no crop was sown as the soil was too wet.

Gas Sampling and Calculations

Emissions of N_2O and CH_4 were measured using closed static chambers. In the spring experiment, emissions were measured on 12 occasions, starting on the day after the application (on days 1, 3, 5, 7, 9, 11, 14, 19, 26, 35, 47, 56 and 67). In the autumn experiment emissions were measured on 11 occasions, starting on the day after application (on day 1, 3, 7, 9, 11, 14, 17, 22, 30, 36 and 45). The sampling regimes were almost identical in spring and autumn up to day 14 and thereafter differed somewhat due to practical limitations. After incorporation of the SS, three iron metal frames (0.525 m × 0.33 m) equipped with a channel-shaped water seal were pushed 0.05 m into the soil at random spots in each plot in spring, in the control plots in autumn and in each of the 1.5 m × 1.5 m randomly placed SS application areas in autumn [37]. At measuring, the channels were filled with water, after which a PVC chamber (internal height 0.215 m) was placed on the frame, so that the air above the soil was enclosed in the

chamber. Gas samples were taken at the time of sealing by inserting a 50-mL plastic syringe into a valve in one chamber of each plot and then from all chambers after 60 min. The samples were injected into gastight 20-mL vials fitted with rubber septa. On the first sampling day, samples were taken at 30, 60, 90 and 120 min from one chamber in each treatment after sealing, to ensure that the increase in concentration in the chambers over time was linear.

In the spring experiment, NH_3 emissions were measured for the first 24 h after application of SS with a dynamic chamber technique [38]. The measurements were made in all three treatments (immediate incorporation, delayed incorporation and control). In one control plot and in all treatment plots, two PVC chambers (0.40 × 0.30 m, internal height 0.18 m) were randomly placed, each with a holder for determining the equilibrium concentration of NH_3, and also with a holder for analysis of ambient air per plot. Passive diffusion samplers (one for concentration measurements and one for measuring laminar boundaries) were placed in each holder. Appropriate exposure times for the diffusion samplers were determined by measuring the instantaneous concentration of NH_3 in the chambers with a hand-held instrument (Kitagawa precision gas detector, Komyo Rikagku Kogyo KK, Tokyo, Japan).

The sampled gas was analysed for N_2O and CH_4 with a gas chromatograph equipped with a capillary column (Elite Plot Q), flame ionisation detector (FID) and electron capture detector (ECD) (Perkin Elmer Clarus 500, Perkin Elmer, Waltham, MA, USA) provided with a headspace auto sampler (TurboMatrix 110, Perkin Elmer). Emissions of N_2O and CH_4 per hour on the sampling day were calculated from the linear increase in concentration over time in the closed chamber. Emissions between two sampling occasions were estimated by multiplying the mean value of the emissions on the two measurement occasions by the number of hours between the occasions. Cumulative values were calculated by adding up the estimated emissions for the entire period. The NH_3 emissions between the two measurement periods were calculated from interpolated values adjusted for the prevailing weather conditions during the interval, according to Malgeryd [27].

Statistical Analysis

All statistical analyses were performed using Statistical Analysis Software (SAS, ver. 9.4, SAS Institute Inc., Cary, NC, USA). Differences between cumulative emissions of the treatments in each experiment were analysed by one-way ANOVA with blocks, followed by pairwise comparisons with t test (PROC GLM). In a randomised complete block design, the variation between blocks is eliminated in treatment comparison, but may contribute

substantially to the standard error of the individual means. Hence, pooled standard errors of treatments are presented. The Pearson correlation coefficients between fluxes of N_2O and CH_4 and soil moisture and soil temperature were calculated and analysed (PROC CORR). For statistically significant differences, the probability value $p < 0.05$ was used throughout.

Results

Environmental and Soil Conditions

Mean air temperature on the day of SS application in the spring experiment (which was also the day for starting NH_3 measurements) was 7.8 °C and the mean soil temperature was 11.4 °C. There was no precipitation and the wind speed was on average 12.5 m s^{-1}. Mean air temperature during the 67-day GHG measuring period was 13.4 (5.4–20.8, min–max) °C (Fig. 1a) and mean soil temperature at 0.02–0.05 m depth was 14.5 (3.1–28.1) °C (Table 2). Cumulative precipitation during the measuring period was 171 mm. Mean air temperature on the day of SS application in the autumn experiment was 8.0 °C, there was no precipitation and the wind speed was on average 1.6 m s^{-1}. Mean air temperature during the 45-day GHG measuring period was 4.5 (−2.3 to 11.4) °C (Fig. 2a) and mean soil temperature at 0.02–0.05 m depth was 3.4 (−1.8 to 10.6) °C (Table 2). Cumulative precipitation during the measuring period was 66 mm.

The characteristics of the soil before SS application are shown in Table 1. At temperatures below 0 °C and on some other occasions, the soil water sensor did not function. Therefore data from those days were excluded from the calculations of average moisture during those periods and thus also from Figs. 1a, and 2a. The soil was rather dry during the spring experiment, with mean vol. WC of 11 (0–25) % (Fig. 1a), whereas during the whole measuring period in autumn the soil was wet, with mean vol. WC of 38 (15–47) % (Fig. 2a).

Sewage Sludge Application

The characteristics of the SS spread in spring and in autumn were similar except for the higher content of TAN in the urea-treated SS used in the spring experiment (Table 1). In spring, the mean application rate of SS for the whole experiment was 7.2 Mg ha^{-1} [with 58 % coefficient of variation (CV); n = 6], which was about half the target application rate of 12 Mg ha^{-1}. The mean actual application rate corresponded to 63 kg P ha^{-1}, 130 kg Tot-N ha^{-1}, 57 kg TAN ha^{-1} and 620 kg Tot-C ha^{-1}. The SS application rate in autumn was 13.7 Mg ha^{-1}, which corresponded to 110 kg P ha^{-1}, 173 kg Tot-N ha^{-1}, 52 kg TAN ha^{-1} and 1030 kg Tot-C ha^{-1}. Since the amount spread on each plot in autumn was individually weighed out and applied manually, the variation in application rates was considered negligible.

Nitrous Oxide Emissions

Daily mean emissions of N_2O over the 67-day sampling period in the spring experiment corresponded to 8.7, 6.2 and 2.3 g N_2O-N ha^{-1} for immediate incorporation, delayed incorporation and the unfertilised control, respectively. Compared with the unfertilised control, cumulative emissions were almost 4-fold and 3-fold as high as for immediate and delayed incorporated SS, respectively (Table 3). However, the differences were not statistically significant. During the measuring period, two emission peaks were observed in the two treatments and the control, the first 9 days after application and the second 35 days after application (Fig. 1b). The emissions from the two SS treatments during the peak on day 9 corresponded to 0.01 and 0.02 kg N_2O-N ha^{-1} and during the peak on day 35 to 0.02 kg N_2O-N ha^{-1} for both immediate and delayed incorporation. Both peaks were preceded by rainfall events totalling 39 mm during the preceding 4 days for the first peak and totalling 40 mm during the preceding 8 days for the second peak. Total net loss of N as N_2O for the spring experiment, calculated by subtracting the emissions from the control treatment, corresponded to 0.32 and 0.20 % of added Tot-N for immediate and delayed incorporation of
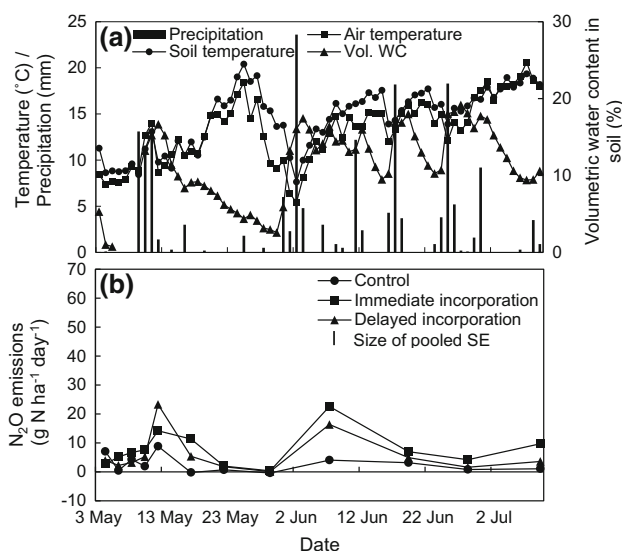

Fig. 1 Precipitation, air and soil temperature, volumetric water content and N_2O emissions after application of sewage sludge treated with urea in spring. Missing values for vol. WC are due to problems with the equipment. All data points for N_2O emissions are mean of three replicates. Pooled standard error (SE): 5.38

Table 2 Mean soil water content, dry bulk density (n = 9, standard error in brackets) and temperature (minimum–maximum in brackets) on application of sewage sludge in spring and autumn, respectively

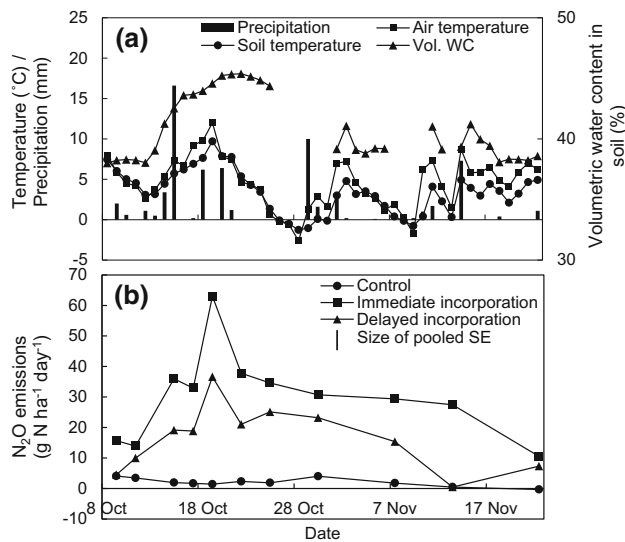| Time	Depth (m)	Water content (% of dry soil)	Dry bulk density (g cm^{-3})	Soil temperature during the GHG measuring period (°C)
Spring	0–0.05	17.5 (4.2)	1.29 (0.09)	14.5 (3.1 to 28.1)
	0.05–0.10	24.6 (0.5)	1.41 (0.10)	
Autumn	0–0.05	31.1 (2.5)	0.79 (0.04)	3.4 (−1.8 to 10.6)
	0.05–0.10	29.2 (1.1)	1.26 (0.07)	

Fig. 2 Precipitation, air and soil temperature, volumetric water content and N_2O emissions after application of sewage sludge in autumn. Missing values for vol. WC are due to soil temperatures below 0 °C, when the measuring equipment did not work. All data points for N_2O emissions are mean of three replicates. Pooled standard error (SE): 7.12

SS, respectively. There was no statistically significant difference between the two treatments. There was also no statistically significant correlation between soil temperature or vol. WC and N_2O emissions.

Daily mean emissions of N_2O over the 45-day long sampling period in the autumn experiment were 29.9, 15.5 and 2.0 g N_2O-N ha^{-1} for immediate incorporation, delayed incorporation and the unfertilised control, respectively. Compared with the unfertilised control, cumulative emissions were 15-fold and 8-fold as high as for immediate and delayed incorporated SS, respectively (Table 3). The difference between the two SS treatments and the control was statistically significant (Table 3), but the difference between the two incorporation times was not statistically significant. The highest emissions of N_2O from the two SS treatments were observed 11 days after application and corresponded to 0.06 and 0.04 kg N_2O-N ha^{-1} day^{-1} for immediate and delayed incorporation, respectively (Fig. 2b). The peak was preceded by a total of 28 mm rain during the preceding 8 days. No emission peak was seen in the unfertilised control. Net loss of N_2O-N in the

measuring period, calculated by subtracting the emissions from the control, corresponded to 0.71 and 0.34 % of added Tot-N for immediate and delayed incorporated of SS, respectively (but with no statistically significant difference between treatments). A statistically significant linear correlation between mean N_2O emissions and mean vol. WC in soil for the whole dataset was seen for both the SS treatment with immediate incorporation (r = 0.84, p < 0.0091) and that with delayed incorporation (r = 0.86, p < 0.0063). No statistically significant correlation was seen between soil temperature and N_2O emissions.

Ammonia Emissions

Total loss of NH_3-N from immediate and delayed incorporation of SS during the first 24 h was 9.2 and 15.4 kg N ha^{-1}, respectively, corresponding to 7.1 and 11.9 % of added Tot-N, respectively, and 16.3 and 27.3 % of added TAN, respectively. There was no statistically significant difference between the two treatments. The emission rate of NH_3 was high during the first 2–3 h after application (5.1 and 8.0 kg NH_3-N ha^{-1} for immediate and delayed incorporation, respectively) and the emission rate thereafter declined in both treatments, but had not fully ceased after 24 h.

Methane Emissions

In the spring experiment, the soil in treatments both with and without SS application generally acted as a sink for CH_4, as shown by negative emissions (Table 3). However, all treatments emitted CH_4 at the first measurement and the treatment with SS incorporated immediately emitted CH_4 on one more occasion. No statistically significant difference was seen between any of the treatments. Furthermore, there was no statistically significant correlation between CH_4 emissions and soil temperature or vol. WC.

The CH_4 emissions fluctuated between negative and positive fluxes during the autumn experiment and cumulative emissions over the measuring period were small (Table 3). There was no statistically significant difference between any of the treatments. Most of the CH_4 was emitted in the two last weeks of the measuring period. Net total CH_4-C loss for the measuring period, calculated by

Table 3 Emissions of N_2O and CH_4 from soil to which mesophilically digested, urea-treated, stored sewage sludge (SS) was applied in spring and mesophilically digested, stored SS was applied in autumn, expressed as cumulative gross and net rates, and as percentage of applied N, TAN and C. The measurements in spring ran for 67 days and those in autumn for 45 days

Time	Treatment	N_2O				CH_4		
		Gross (kg N ha^{-1})[1]	Net (kg N ha^{-1})[2]	(% of Tot-N)[2]	(% of TAN)[2]	Gross (kg C ha^{-1})	Net (kg C ha^{-1})[2]	(% of Tot-C)[2]
Spring	Control	0.15	NR	NR	NR	−0.097	NR	NR
	Immediate incorporation	0.57	0.42	0.32[3]	0.74	−0.116	−0.020	−0.00[3]
	Delayed incorporation	0.41	0.26	0.20[3]	0.46	−0.113	−0.016	−0.00[3]
Autumn	Control	0.09[a]	NR	NR	NR	0.003	NR	NR
	Immediate incorporation	1.31[b]	1.23	0.71	2.38	0.009	0.006	0.00
	Delayed incorporation	0.68[b]	0.60	0.34	1.16	0.016	0.013	0.00

NR not relevant

[1] Statistical analyses of emissions of N_2O-N are based on log transformed data

[2] Cumulative emissions minus emissions from the control treatment

[3] Uncertain values because of large variation in SS application rate

[a, b] Mean values with different letters within columns and occasions are significantly different ($p < 0.05$)

subtracting the emissions from the control, was negligible for both SS treatments (Table 3). No statistically significant correlation was seen between CH_4 emissions and soil temperature or vol. WC.

Discussion

The results showed that N_2O emissions from SS applied to soil at rates based on the maximum dose of P permitted by Swedish environmental regulations (110 kg P ha^{-1} during a 5-year period) were relatively low, regardless of time of application during the growing season or time from application to incorporation. Emissions of CH_4 were negligible or even negative, meaning that the field soil at the study site acts as a sink for CH_4.

Nitrous Oxide Emissions

The lowest N_2O emissions were observed in spring and a likely reason for this was the drier soil, as suggested by the statistically significant linear correlation between vol. WC and N_2O emissions observed in the autumn. Similar emission patterns have been reported previously [21] where the release of N_2O from soil treated with dewatered SS was higher during wetter periods of the year than in drier periods. A correlation between soil moisture and N_2O emissions was also reported by Perälä et al. [22], who showed that just a small increase in soil moisture could potentially increase N_2O production in soil treated with organic fertiliser.

The observed peaks in N_2O emissions were preceded by rain in both seasons. Other studies on application of SS [14] and of cattle slurry [24] have also shown that the fluxes of N_2O increase after precipitation. Scott et al. [14] attributed several of the emission peaks in their field study to rainfall causing temporary partly waterlogged soils. The absence of further emissions peaks in autumn in the present study, despite two more occasions of fairly high rainfall, could be explained by the simultaneously low temperature, as proposed by Scott et al. [14]. The absence of emission peaks in spring other than during the first period of the experiment, despite continuous rainfall, are not easily explained by weather factors, but are more likely a consequence of depletion of available mineral N by gaseous losses and N uptake by plants [25, 26]. Nitrous oxide production from N mineralised from Org-N was probably only marginally accounted for, as the mineralisation during the study periods, 67 days in spring and 45 days in autumn, probably were small. However, in both seasons studied, most of the N_2O was emitted early in the experiments. It can be assumed that such early emissions of N_2O are related to easily degradable compounds in the SS creating beneficial conditions for denitrification. However, since the concentrations of easily degradable compounds were not measured, we can only make assumptions about their actual involvement. Nevertheless, these assumptions are supported by previous studies showing that most N_2O is emitted in the first 1–2 months after application of organic fertiliser [13, 15, 24].

The lower emissions of N_2O observed in spring compared with autumn could partly be explained by the spring barley sown in spring, whereas in autumn the SS was spread on stubble and no crop was sown. Jarecki et al. [26] and Parkin et al. [25] demonstrated in 32- to 40-day laboratory-scale experiments that soil cropped with rye and

fertilised with liquid pig manure equivalent to approximately 190 kg N ha^{-1} emitted less N_2O than when no crop was grown. This demonstrates that a growing crop withdraws N from the microbial processes producing N_2O, an effect also seen for lower fertiliser doses (30, 75 and 180 kg N ha^{-1}) in those studies.

Another reason for the lower emissions of N_2O in spring compared with autumn might be that the pathogen reducing effect of the ammonia from the added urea [34] also affected the microbes involved in N_2O production negatively.

It is possible that there would have been slightly higher cumulative emissions from the autumn application if the measuring period had been extended by 22 days to match the length of the measuring period in spring. However, this would have increased the difference between emissions in spring and in autumn. Moreover, during the days after the last sampling occasion in autumn the soil temperature decreased to below zero and remained at freezing point at least until the beginning of April in the next year (data not shown). Hence the N_2O emissions during that period were most likely very low.

With the exception of a few occasions in spring, measured N_2O emissions from the treatment with immediate incorporation were higher than those from the delayed incorporation. Delayed incorporation reduced cumulative net emissions by 38 % in spring and 50 % in autumn (although the differences were not statistically significant in either season).

The lack of significant differences between treatments could in spring partly be explained by the uneven application, with CV 58 % for sampling areas of 0.25 m^2. In the same time, the rather high number, 3, of chambers per small plot should to some extent compensate for the uneven application. However, since the CV of the application was low in autumn, this is not the only explanation.

Few, if any, other studies have previously investigated the effect of immediate versus delayed incorporation of SS. However, our results are in agreement with those of Webb et al. [32], who observed a 30 % reduction in cumulative N_2O emissions in a 46-day measuring period after application of pig manure (DM content 25.5 %) to soil at a rate corresponding to 180 kg Tot-N ha^{-1} (30 kg TAN ha^{-1}) when it was incorporated within 4 h compared with immediate incorporation. However, on repeating the experiment in the following year, they found that emissions of N_2O from immediate and delayed incorporation did not differ [32]. Results from studies where the fertiliser had been either incorporated or left on the surface are contradictory. Thorman et al. [29] reported lower emissions from pig manure (DM content 20.4 %) applied at a rate of 236 kg Tot-N ha^{-1} (4.9 kg TAN ha^{-1}) when left on the surface than when incorporated within 4 h, but achieved

the opposite results with cattle manure [application rate 185 kg Tot-N ha^{-1} (0.4 kg TAN ha^{-1})]. This was also found by Webb et al. [32] [application rate 110–145 kg Tot-N ha^{-1} (8–10.5 kg TAN ha^{-1})]. Various studies comparing incorporation with surface application of animal slurry have demonstrated higher emissions of N_2O from incorporation [23, 24, 28]. Other studies on animal slurry have not shown any statistically significant differences between application methods [30, 31]. It is reasonable to assume that incorporation of N-rich organic fertiliser into the soil leads to both altered amounts of N in the soil matrix and altered micro-environment, affecting the microbes transforming N to N_2O [23]. Incorporation increases the number of places where NH_4^+ and easily available organic matter are found in direct proximity, which can stimulate N_2O production as these are sources of energy for the microbes involved in nitrification and denitrification [23, 37]. Incorporation of SS into the soil might also create anaerobic zones [39], and if these expand, both nitrifying and denitrifying microbes can be active at the same time, implying potential production of large amounts of N_2O [40]. However, since the SS in the present study for which incorporation was delayed only stayed on the soil surface for 4 h, it is unlikely that the mechanisms mentioned above are the only reason for the observed difference between the treatments. It is difficult to find any obvious reasons for the difference between treatments, especially since the difference in NH_3 emissions was very low.

The emission rate of N_2O in the autumn experiment in the present study resulted in similar daily emissions as from dewatered and air-dried SS (17.1 g N_2O-N ha^{-1}) applied to arable land at a rate of 150 kg Tot-N ha^{-1} (5.8 kg TAN ha^{-1}) in a study by Chiaradia et al. [16]. Ambus et al. [15] found no differences in N_2O emissions between a soil fertilised with the comparatively low application rate of 81 kg Tot-N ha^{-1} (TAN not given) SS and a soil without SS application. In contrast, Scott et al. [14] reported an emission factor of 1 % for a 208-day period for N_2O from digested and dewatered SS applied on arable land, which is 1.4-fold and 5-fold as high as the highest and lowest results in the present study (immediate incorporation of SS in the autumn and delayed incorporation of SS in the spring, respectively). The SS used by Scott et al. [14] was much drier (66 % DM) than in the present study, which could imply lower emissions. However, since their study concerned SS disposal on land rather than use of SS for fertilisation purposes, the Tot-N application rate was 2500 kg Tot-N ha^{-1} (91 kg TAN ha^{-1}), which is 14-fold and 19-fold, respectively, as high as in spring in the present study and also much greater than the crop requirement. Similarly, Mosier et al. [13] reported an emission factor of 0.8 and 1 %, respectively, for a 155-day period for N_2O

from SS applied to fields at an application rate of 290 and 1440 kg Tot-N ha^{-1} (TAN not given), which is also much higher than in the present study. Higher Tot-N application rates have previously been shown to lead to higher emissions of N_2O of applied N from animal slurry applied to arable land [23, 33] and are one explanation for the different results in the studies by Ambus et al. [15], Scott et al. [14] and Mosier et al. [13] compared with the present study.

de Urzedo et al. [19] reported an emission factor of 0.95 % for a 172-day period for N_2O from dewatered SS applied to arable land, which is higher than in the present study, despite the low application rate of 65 kg Tot-N ha^{-1} (2.05 kg TAN ha^{-1}). On the contrary, despite high application rates [300 or 600 kg Tot-N ha^{-1} year^{-1} for 7 years (3.7 or 7.4 kg TAN ha^{-1})], Pitombo et al. [21] reported lower daily emissions (2.1 and −0.07 g N_2O-N ha^{-1} day^{-1} in the wet and dry season, respectively, with the lower application rate; 4.7 and −0.13 g N_2O-N ha^{-1} day^{-1} respectively, with the higher application rate) than in the present study. One explanation for this may be that 2 years elapsed between last SS application and start of GHG measurements. Studies conducted in greenhouses in which digested SS was applied to soil at a rate of 150 kg Tot-N ha^{-1} (15.4 and 32.9 kg TAN ha^{-1} for respective study), which is a similar N rate to that in the present study, reported much lower mean daily emissions ranging from 0.01 to 0.8 g N_2O-N ha^{-1} day^{-1} [18, 20]. However, daily emission rates in a similar greenhouse study [17] with digested SS at a lower application rate [80 kg Tot-N ha^{-1} (TAN not given)] were similar (8.0 g N_2O-N ha^{-1} day^{-1}) to those in the spring experiment in the present study. This variation in results indicates that factors other than those discussed above, such as soil properties, climate factors, SS application methods and SS properties also influence the magnitude of the N_2O emissions.

Ammonia Emissions

The NH_3 loss as a percentage of added N from immediate incorporation of SS with urea addition in spring (7.1 %) was lower than that reported from manure (21 %) and farmyard manure (FYM) with urea (14 %) in a review of NH_3 emissions by FAO and IFA [41]. However, the NH_3 loss as a percentage of added TAN from immediate incorporation (16.3 %) was slightly higher than the Swedish default value for immediate incorporation of solid manure (15 % of added TAN) [42]. The NH_3 loss from delayed incorporated SS in spring (11.9 % of added Tot-N and 27.3 % of added TAN) was also slightly lower than that for manure and FYM with urea presented by FAO and IFA [41], and somewhat lower than the Swedish default value (33 % of added TAN) for incorporation of solid

manure after 4 h [42]. Other studies report emissions of NH_3 from SS application of 60 % of 150 kg TAN ha^{-1} applied [43], or 0–32 % of NH_3-N applied, interpreted as TAN applied [44]. The relatively low emission rate of NH_3 in the present study could partly be explained by rather low air temperatures during the measuring period compared to other studies with SS [43, 44], but also by the emissions only being measured for 24 h and the emission had not fully ceased at this time. It was interesting that the difference between NH_3 emissions for immediate and delayed incorporation was so small, since it has previously been shown that incorporation or injection of organic fertilisers into soil are effective measures to reduce NH_3 losses [24, 28]. The small difference in NH_3 losses between treatments are partly explained by the variation in application rate (CV = 58 %).

Methane Emissions

Cumulative emissions of CH_4 were either close to zero or negative in both spring and autumn in this study. Previous studies of SS application to soil have reported similar effects [15, 18, 20, 21]. In spring, almost all CH_4 emissions occurred early in the experiment and after this, only negative fluxes were observed. Since CH_4 production only takes place under strictly anaerobic conditions, some anaerobic microsites might have developed at the time of application, for example in lumps of SS. Alternatively, the CH_4 emitted immediately after application of SS may have been produced through methanogenic activity during the preceding storage, as discussed by Rodhe et al. [37]. In the autumn, however, most of the CH_4 emissions occurred on the second last measuring occasion. No recorded climate data could explain this peak.

In general, aerated arable soils (not subjected to flooding) are considered sinks of CH_4 [10, 11]. The higher CH_4 emissions in autumn compared with spring, though not statistically significant, might partly be explained by the higher soil moisture in the autumn giving a higher proportion of water-filled soil pores, leading to reduced CH_4 oxidising capacity of the soil [45]. However, in autumn the soil in the present study had lower dry bulk density than in spring, partly because of more crop residues in the soil after harvest. Thus, the results in the present study contradict those of Smith et al. [45], who found that lower dry bulk density increased CH_4 oxidation ability. In the present study, the trend for higher CH_4 oxidation capacity in spring occurred despite higher dry bulk density, indicating that the effect of the lower soil moisture in spring was larger than the effect of the higher dry bulk density.

Negligible emissions can be explained by simultaneous occurrence of methanogenic and methanotrophic activity

reducing or eliminating the release of CH_4 to the atmosphere [46]. Another explanation could be the low soil temperatures in both the spring and autumn experiments decreasing methanogenic activity and other microbial activity in the chain leading to CH_4 formation [10]. The fluxes of CH_4 from the control soil varied seasonally, with more occasions of positive fluxes in the autumn compared with spring. Such variation was also reported for long-term measurements by Smith et al. [45], who demonstrated that the ability of the soil to absorb CH_4 changes over the year.

Practical Applications

The impact that urea addition seemed to have on the physical properties of the SS needs to be taken into account when using urea-treated SS as fertiliser. Scott et al. [14] also reported problems with variations in measured emissions within treatments and attributed this to possible uneven distribution of fertiliser, but also to spatial variation in microbial activity in the soil.

Nitrogen uptake is more efficient in spring and, based on the findings in this study, there are indications that conditions more common in spring than in autumn (e.g. drier soils and less precipitation) are also beneficial in terms of GHG emissions reduction. Application of SS in Sweden is more commonly carried out in autumn than in spring, mostly due to the risk of soil compaction in spring, but also because of SS being considered a P fertiliser with low concentration of mineral N. It has been shown that storing SS for long periods results in large GHG emissions during storage [47]. Sewage sludge application in spring would shorten the storage period and thereby reduce emissions from storage. Incorporation or injection of SS after application might involve increased emissions of N_2O, as indicated by the consistent tendency in this study. However, these application measures are well known to reduce NH_3 emissions and a reduction in NH_3 also reduces the indirect N_2O emissions [24, 28]. Similar goal conflicts between N_2O and NH_3 emissions for cattle slurry have been shown by Wulf et al. [24] and Rodhe et al. [37] and for pig slurry by Thomsen et al. [28].

Conclusions

In general, emissions of N_2O from SS applied to arable land at a rate based on the maximum permissible P level according to Swedish environmental regulations were moderate and emissions of CH_4 negligible. Soil moisture was an important controlling variable for N_2O emissions, which tended to be higher in autumn than in spring. There was a consistent tendency for precipitation to result in emissions peaks. There was no statistically significant difference in emissions of N_2O, CH_4 or NH_3 between incorporating SS immediately and delaying this by 4 h. However, there was a consistent tendency for delayed incorporation to result in lower emissions of N_2O and higher emissions of NH_3. Emissions of CH_4 were very low or negative, indicating CH_4 oxidising capacity in the soil.

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