Emissions of greenhouse gases (N_2O, CH_4 and NH_3) after land application of urea-sanitised sewage sludge under Swedish conditions

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Abstract
According to the Swedish Environmental Quality Objectives, 60% of the phosphorus in wastewater should be recycled to land by 2015. Legislation has been proposed requiring that sewage sludge be sanitised prior to land application, with the minimum sanitisation standard being storage for one year. The sanitisation requirement can also be met by e.g. adding urea. A field experiment was conducted to quantify CH_4, N_2O and NH_3 emissions after land application of dewatered sewage sludge treated with urea, either immediately incorporated into the soil or four hours after application. Delayed incorporation seemed to lower N_2O emissions compared with immediate incorporation, possibly due to higher NH_3 losses observed in the experiment. Cumulative CH_4 emissions were negative, indicating oxidation of CH_4.

Introduction
One target under the Swedish Environmental Quality Objectives is for 60% of the phosphorus (P) in wastewater to be recycled to land by 2015. In 2010, about 204 000 tonnes of sewage sludge were produced in Sweden and 25% of this was applied to arable land [1]. Legislation has been proposed requiring that sewage sludge be sanitised prior to land application, with the minimum sanitisation standard being storage for one year. The sanitisation requirement can also be met by e.g. thermophilic digestion or by adding urea. The main greenhouse gas (GHG) emitted from fertilised soil is nitrous oxide (N_2O), but methane (CH_4) is also emitted, mainly directly after application of organic fertiliser. A common practice to reduce ammonia (NH_3) emissions and odour from manure is to incorporate it into the soil at application, or as soon as possible afterwards. Temperature and precipitation influence the magnitude of the GHG emissions, and both diurnal and seasonal variations occur [2]. The objective of this study was to quantify GHG emissions after applying sewage sludge to arable land in spring before sowing and to analyse the GHG reduction potential of immediate compared with delayed incorporation. The hypothesis was that emissions can be minimised by applying an appropriate spreading strategy for the type of sludge used.

Materials and Methods
In a field experiment, GHG emissions were quantified after land application of dewatered sewage sludge treated with urea, either immediately incorporated into the soil or incorporated four hours after application. Unfertilised soil was used as the control.

Sewage sludge
The sewage sludge used in the experiment was mesophilically digested and dewatered sewage sludge sanitised by adding urea (1.5% by weight) and stored covered for five months. At the start of the experiment, the sludge was analysed for content of dry matter (DM), total nitrogen (Tot-N), organic nitrogen (Org-N), ammonium nitrogen (NH_4-N), total carbon (Tot-C), C/N-ratio (Tot-C/Tot-N) and total phosphorus (Tot-P).

Field site and soil characteristics
The study site was located 8.2 km west of Uppsala (59˚53'N, 17˚32'E). The sludge was applied on arable land on 3 May 2012. Spring application was examined to make use of the elevated content of plant-available nitrogen from the urea. Before spreading, cylinder samples (50 mm high and 72 mm in diameter) were collected for determination of soil bulk density and soil moisture. The samples were taken at 0-0.05 m and 0.05-0.10 m depth, respectively, at three locations per block, giving a total of 18 samples. A composite sample from the soil (15 subsamples) was taken at 0-0.2 m depth for texture

analysis. During the experiment, soil temperature at 0.02-0.05 m depth was recorded hourly using Tiny Tag dataloggers (Intab AB, Stenkullen, Sweden) and the volumetric moisture content (vol. MC) in the topsoil was recorded every 10 minute using a Theta Probe ML2x sensor (Delta-T Devices Ltd, Cambridge, UK) [3].

Experimental set-up and design

The experiment was organised in a randomised complete block design with three blocks. The six plots treated with sludge were 6 m x 12 m each and the three control plots 3 m x 12 m each. The target rate for sewage sludge application was 13 tonnes ha^{-1}. The sewage sludge was applied using a JF ST 9500 solid manure spreader with two horizontal beaters equipped with a hydraulically driven bottom conveyor and a front wall for more uniform longitudinal distribution of the manure. The sludge was incorporated into the upper soil layer either immediately or four hours after application, using a harrow. Immediate incorporation was chosen as it is a well-documented and efficient method for reducing NH_3 emissions, and a four-hour delay as this is the longest time permitted before incorporation of fertilisers containing urea according to current regulations [4]. After incorporation, spring barley was sown in the field.

Weather

Meteorological data for the experimental period were obtained from a weather station operated by the Department of Earth Sciences at Uppsala University [5]. The weather station is situated in north-west Uppsala, approximately 8 km from the study site. During measurement of NH_3 emissions, a stationary weather station at the study site was used (data not shown).

Gas measurements

Emissions of N_2O and CH_4 were measured by a closed chamber technique on 12 occasions over the 67 days following sludge application (on days 1, 3, 5, 7, 9, 14, 19, 26, 35, 47, 56 and 67 days), i.e. with more frequent sampling initially [6]. In each of the nine plots, three sampling frames (0.525 m x 0.33 m) were positioned randomly and pressed 0.05 m into the ground. Gas samples were collected from one chamber in each plot at the time of closing and then in all chambers after one hour. The samples were analysed for CH_4 and N_2O using a GC (HP 6890, Hewlett Packard). Total cumulative emissions and daily emissions rate of N_2O and CH_4 were determined for each treatment. NH_3 emissions were measured during the first days after application with an equilibrium concentration method that combines the dynamic chamber technique with passive diffusion sampling to measure the difference in NH_3 concentration between soil and air [7].

Results and discussion

Sewage sludge

The chemical characteristics of the urea-treated sewage sludge at the start of the storage period are shown in Table 1.

Table 1. Chemical characteristics of the urea-treated sewage sludge at the start of the storage period

DM (%)	Tot-N	Org-N	NH_4-N (kg ton^{-1})	Tot-C	C/N-ratio	Tot-P
26.9	18.0	10.2	7.9	85.8	4.8	8.7

Application and incorporation

Due to technical problems during application, the sludge was not as evenly distributed in the field as desired, which may have caused a variation in fluxes between the sampling chambers.

Soil characterisation

The moisture content and bulk density in the soil prior to sludge application were higher in the 0-5 cm layer than in the 5-10 cm layer (Table 2).

Table 2. Mean soil moisture and bulk density in the soil (n=9, standard deviation in brackets)

Measuring depth (cm)	Water content (% of dry soil)	Bulk density (g cm^{-3})
0-5	17.5 (4.23)	1.29 (0.09)
5-10	24.6 (0.56)	1.41 (0.10)

Texture analysis showed that the soil was a silty clay (42% clay, 48% silt, 10% sand).

Weather

The mean temperature for the 67 days of measurements was 13.4˚C (Figure 1). Total precipitation for the period was 170.0 mm [5].

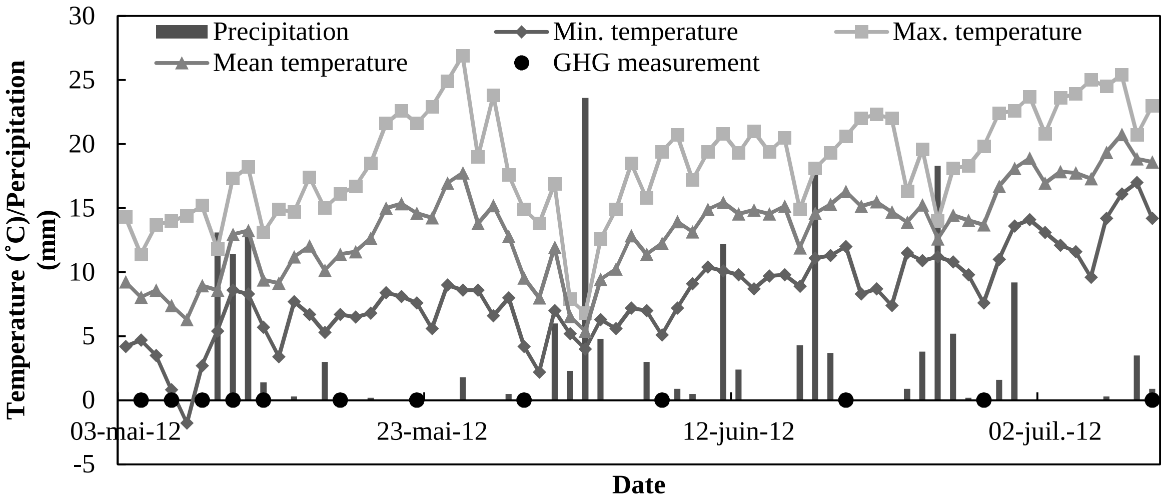

Figure 1. Precipitation (bars) and mean, minimum and maximum temperatures in Uppsala during the study period. GHG measurement occasions are marked with closed circles (●).

Gas measurements

There were two occasions with high N_2O fluxes (day 5 and 9, data not shown), both following precipitation some days earlier (Figure 1). Moreover, on a few occasions the flux of N_2O was higher for the sludge incorporated after four hours than for the immediately incorporated sludge (data not shown. However, in cumulative terms immediate incorporation of sludge led to higher overall emissions of N_2O than delayed incorporation (Table 3). On the two first sampling occasions, positive fluxes of CH_4 were observed from all three treatments (data not shown). At sampling at the end of June, positive fluxes of CH_4 also occurred from the treatment with sludge immediately incorporated. However, the generally negative values of CH_4 fluxes in all treatments showed that the soil generally acted as a sink for CH_4, both without and with sludge applied (Table 3).

Table 3. Total cumulative emissions (66 days) and daily mean emission rates of N_2O and CH_4

Treatment	Cum. N_2O emissions (g N_2O-N ha^{-1})	Daily mean N_2O emissions (g N_2O-N ha^{-1})	Cum. CH_4 emissions (g CH_4-C ha^{-1})	Daily mean CH_4 emissions (g CH_4-C ha^{-1})
C	204	3.09	-39.1	-0.59
I	609	9.23	-50.6	-0.77
D	470	7.12	-52.7	-0.80

C = control, I = immediate incorporation and D = delayed incorporation

Daily mean N_2O emission rate, i.e. total cumulative N_2O emissions divided by number of measuring days, was 9.23 g N_2O-N ha^{-1} for the immediately incorporated sludge and 7.12 g N_2O-N ha^{-1} for the sludge incorporated after four hours (Table 3). The daily mean emission rate of CH_4 was negative for all treatments (-0.59, -0.77 and -0.80 g CH_4-C ha^{-1} for the control, immediate incorporation and delayed incorporation, respectively) (Table 3).

Emissions of NH_3 were measured for 48 hours after which the emissions stalled. Cumulative emissions of NH_3 during this period, calculated as loss of ammonium nitrogen (NH_4-N) were higher for the sludge incorporated after four hours than for the sludge incorporated immediately after application (Table 4).

Table 4. Total nitrogen application and losses of ammonia after immediate or delayed incorporation of urea-treated sewage sludge applied to arable soil in spring

	Tot-N applied (kg ha^{-1})	NH_4-N applied (kg ha^{-1})	NH_4-N loss (kg ha^{-1})
Immediate incorporation	234	102	13.3
Incorporation after 4 h	234	102	15.4

Conclusions and perspectives

Delayed incorporation seemed to lower N_2O emissions compared with immediate incorporation, possibly due to the higher NH_3 losses measured in the experiment. Emissions of NH_3 decrease the amount of mineral N substrate available for ammonium-oxidising bacteria, which are known to produce N_2O under semi-moist conditions such as those in this study. Cumulative CH_4 emissions were negative, indicating oxidation of CH_4. Emissions of CH_4 from most arable soils are low and CH_4 is produced mainly under extreme anaerobic conditions such as those in paddy soils. Methane oxidising bacteria are common in arable soil ecosystems and hence CH_4 is consumed wherever CH_4 and oxygen are present together.

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