Sewage sludge as fertiliser – environmental assessment of storage and land application options
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ABSTRACT

Sewage sludge (SS) contains beneficial plant nutrients and organic matter, and therefore application of SS on agricultural land helps close nutrient loops. However, spreading operations are restricted to certain seasons and hence the SS needs to be stored. Storage and land application of SS are both potential sources of greenhouse gases and ammonia, leading to global warming, acidification and eutrophication. Covering the stored SS, treating it with urea and choosing the correct time for land application all have the potential to reduce emissions from the system. Using life cycle assessment (LCA), this study compares storage and land application options of SS in terms of global warming potential (GWP), acidification potential, eutrophication potential and primary energy use. The system with covered storage has the lowest impact of all categories. Systems with autumn application are preferable to spring application for all impact categories but, when nitrate leaching is considered, spring application is preferable in terms of eutrophication and primary energy use and, for some SS treatments, GWP. Ammonia addition reduces nitrous oxide and ammonia emissions during storage, but increases these emissions after land application. Storage duration has a large impact on GWP, while amount of chemical nitrogen fertiliser substituted has a large impact on primary energy use.

Key words | acidification, biosolids, eutrophication, GWP, LCA, primary energy use

INTRODUCTION

In agriculture, sustainable development includes seeking to close plant nutrient loops. The single largest flow of phosphorus (P) from Swedish agriculture is associated with products for human consumption, both food and fibre. A large proportion of this P ends up in sewage sludge (SS), a potential organic fertiliser also rich in plant nutrients such as nitrogen (N) and sulphur. In addition, SS contains organic matter which, when applied to arable land, can improve soil structure and increase soil carbon content. Approximately 12,970,000 Mg dry matter (DM) of SS are produced in the European Union annually, of which approximately 38% is applied to arable land (Eurostat 2012). In Sweden, 200,000 Mg DM are produced annually, of which approximately 23% is applied to arable land (Eurostat 2012).

Digested SS used as fertiliser replaces chemical fertilisers and thereby avoids energy- and resource-demanding manufacture of chemical fertilisers with its associated greenhouse gas emissions. The digestion (which is often anaerobic) also results in renewable energy in the form of biogas, which can replace fossil resources and thereby reduce emissions that lead to global warming. Recycling of N and P can contribute to reducing global use of N and P chemical fertilisers by 50–60%, as recommended by Steffen et al. (2013), to prevent biogeochemical flows of these elements exceeding their boundaries for safe planetary development. Sweden had a longstanding national goal of recycling 60% of the P in sewage to agricultural land by 2015 (Swedish Government 2009). This goal has now expired, and a new goal of recycling 40% of the P and 10% of the N in SS to arable land has been proposed by the Swedish Environmental Protection Agency, but has not yet been agreed by the Swedish government.

According to the Intergovernmental Panel on Climate Change (IPCC), the waste and wastewater industry contributed approximately 3% to global anthropogenic emissions of greenhouse gases (GHGs) in 2010 (IPCC 2013). This was largely due to emissions of methane (CH₄) and nitrous oxide...
(\text{N}_2\text{O})$ during the different steps of waste, wastewater and SS management, the global warming potential (GWP) of which corresponded to 25 and 298 carbon dioxide equivalents (\text{CO}_2\text{-eq}) respectively in a 100-year perspective (IPCC 2007). Emissions of mineral $\text{N}$, i.e. ammonium ($\text{NH}_4^+$), ammonia ($\text{NH}_3$) and nitrate ($\text{NO}_3^-$), from wastewater treatment and SS management also lead to increased acidification potential, eutrophication potential and indirect $\text{N}_2\text{O}$ production as well.

In Sweden, SS (and solid manure) is generally applied to arable land in autumn, partly due to the risk of soil compaction in spring and partly because SS is considered a P fertiliser with a low mineral $\text{N}$ concentration. However, applying SS only in autumn means that most of it has to be stored for rather a long time before use. During storage, GHGs such as $\text{CH}_4$, $\text{N}_2\text{O}$, and $\text{CO}_2$ (mainly biogenic) as well as $\text{NH}_3$ are emitted due to microbiological and physico-chemical transformations, causing direct and indirect greenhouse effects and high acidification and eutrophication potential. The magnitude of emissions is affected by the duration and conditions of storage. In general, more aerobic storage decreases $\text{CH}_4$ emissions, but can increase $\text{N}_2\text{O}$ emissions (Willén et al. 2016a). Emissions of GHG are reduced at lower temperatures (Sommer et al. 2004; Willén et al. 2016a). Ammonia treatment of SS by the addition of urea can be used to reduce its content of pathogens. This treatment also eliminates $\text{N}_2\text{O}$ emissions and significantly reduces $\text{CH}_4$ emissions (Willén et al. 2016a), but increases the risk of $\text{NH}_3$ emissions, necessitating mitigation measures such as covering the stored SS.

After application of SS to soil, emissions of $\text{N}_2\text{O}$, $\text{CH}_4$ and $\text{NH}_3$ may continue, depending on the environmental conditions and normal soil microbial activity (Willén et al. 2016b). On soils with a low clay content in particular, there is also a potential risk of $\text{N}$ leaching as $\text{NO}_3^-$ during autumn when there is no growing crop to take up nutrients (Aronsson & Torstensson 2004). In Sweden, the weather is normally warmer in spring and the soil drier than in autumn, and thus on average the $\text{NH}_3$ emissions after spring application of organic fertilisers are higher than after autumn application (Karlsson & Rodhe 2002). On the other hand, after just a few weeks in spring, the growing crop starts to take up mineral $\text{N}$, thus decreasing the potential for $\text{N}_2\text{O}$ emissions and $\text{NO}_3^-$ losses. Furthermore, by spreading SS in spring, the amount of SS stored during summer is decreased, lowering the emissions of GHG from storage.

Previous studies on SS management have focused on application techniques (Pradel et al. 2015) or compared end-use scenarios that often exclude SS storage (for example, Suh & Rousseaux 2002). However, in a life cycle assessment (LCA) study on SS management that included both storage and land application, Johansson et al. (2008) clearly demonstrated the importance of including both these operations for accurate results. Studies on landfilling of SS have reported that $\text{CH}_4$ emissions can contribute considerably to GWP (Suh & Rousseaux 2002; Houillon & Jolliet 2005), justifying avoidance of landfilling, for example by reusing SS as fertiliser. This raises the question of how SS as a fertiliser should be managed in order to minimise its negative environmental impacts.

The LCA approach is well suited for assessing the environmental impact of different services and products, since it provides quantitative information on resource use and environmental emissions of the systems investigated. The objective of this study is to assess the GWP, acidification and eutrophication potential, and primary energy use for the combination of storage and land application of SS using LCA methodology. The effects of combinations of different storage techniques, storage durations and land application times were investigated. The overall aim was to provide better decision support on management of SS as a fertiliser, in order to lower its environmental impact.

METHODOLOGY

LCA is a quantitative method for assessing a system’s environmental impact during its life cycle. An LCA is divided into four phases: goal and scope, inventory analysis, impact assessment and interpretation. The procedure is iterative, and new observations made along the way are incorporated and can influence all phases of the work. The LCA methodology used here is as specified in ISO 14040 (ISO 2006).

Goal and scope

The goal of this study was to assess the environmental impact of different combined strategies for storage and land application of SS. Different SS storage techniques and application times were compared for a location in south-east Sweden with a mean annual temperature of 8°C and mean annual precipitation of 650 mm. Most of the soils in the study region have a high clay content. In all scenarios, the SS was digested and dewatered, since this is the dominant form of SS management in Nordic countries. The following storage treatments were included...
in the study: storage of SS without a cover (S), storage of SS with a cover (CS), and storage of SS with a cover and treated with NH$_3$ during storage (CAS). Three application times were tested for all three SS treatments: all SS applied in autumn (July–October), all SS applied in spring (mid-April–mid-May), and SS applied in both autumn (80% of SS) and spring (20% of SS). The latter scenario was chosen because approximately 80% of the SS applied to arable land managed by one of the largest distributors of SS in Sweden was applied in autumn in 2015 (Wigh, personal communication). Each SS storage treatment was combined with each land application time, giving a total of nine scenarios.

Functional unit

The functional unit (FU) used in the study was the amount of digested and dewatered SS containing P replacing 1 kg of chemical P fertiliser. It was assumed that 60% of total P content in the SS replaced chemical P fertiliser (Foley et al. 2009).

System boundaries

The system studied started with the SS leaving the digestion plant to be loaded onto trucks for transportation to the storage facility (Figure 1). The emissions and primary energy use associated with all transport, storage and land application were included. Emissions from storage and after land application included N$_2$O, CH$_4$, NH$_3$ and indirect N$_2$O emissions from emitted NH$_3$. It has been suggested that not all the carbon in domestic wastewater is biogenic (Griffith et al. 2009). However, for simplicity and in accordance with IPCC (IPCC 2006), it was assumed here that 100% of the organic carbon in the raw sewage was biogenic, and hence CO$_2$ was not included. In the treatments where the SS was stored under cover, manufacture of the plastic cover material was included, and in the NH$_3$ treatment, manufacture of the urea used as a source of NH$_3$ was included. However, disposal and recycling of used covers was not included, since an initial estimate showed this to have very little impact on the system. Potential nutrient leaching during SS storage was also not included in the analysis, since no such data are available for SS. Moreover, potential P and N leaching and N immobilisation after land application of SS were not included in the study as, given the high clay content, the NO$_3$ leaching potential is close to zero. Potential leaching of NO$_3$ after land application in autumn was included in a sensitivity analysis, since it occurs from some soils. The environmental impact from the manufacture of equipment with a long life, such as vehicles, machinery etc., is often small in relation to the impacts from operations (Johansson et al. 2008). Hence, emissions from manufacturing were excluded from the LCA and only emissions from operations were considered.

Potential substitution of the avoided manufacture and transport of chemical fertiliser were included (Figure 1). Avoided spreading of chemical fertilisers was not included, however, since it has a very low impact (Johansson et al. 2008) and as the number of spreading operations for chemical fertilisers would remain unchanged even if the amount and composition of fertiliser changed, since spreading of chemical fertiliser and sowing were assumed to be combined, i.e. the chemical fertiliser is spread by special coulters in the combined sowing machine.

System description and data used

It was assumed that digested and dewatered SS was transported by truck from the wastewater treatment plant to the storage facility. For the CAS treatment, urea was assumed...
to be mixed into the SS before storage. Ammonia treatment by urea addition was included as a sanitisation measure, but it also increases the value of SS as a fertiliser by the addition of N to the SS. Thereafter, for treatments CS and CAS, the SS was covered with a polyethylene sheet and stored. After the set storage period, the SS was loaded into a tractor-drawn spreader and applied to agricultural land. Emissions from storage and after land application of SS are presented in Tables 2 and 3, respectively, and detailed in the inventory data section. The use of SS replaced the use of chemical fertilisers, modelled as triple super phosphate (TSP) and ammonium nitrate (AN). These were both assumed to be produced by Yara in Finland, and thus their manufacture and transport, as well as avoided emissions of N2O after land application of avoided chemical AN, were accounted for in the calculations.

The system was divided into three subsystems: transport, storage and land application (Figure 1). The first subsystem included primary energy use and emissions from fuel combustion during transport of SS, while the second included emissions during storage and primary energy use and emissions during manufacture of the cover material and urea. The third subsystem included emissions from soil after land application of SS, the primary energy use and emissions from fuel combustion by the tractor used for spreading, and the primary energy use and emissions from avoided manufacture and transport and emissions after land application of replaced chemical fertilisers.

Impact categories

The impact categories studied were GWP, acidification potential, eutrophication potential and primary energy use. GWP was calculated in a 100-year perspective (GWP100) and expressed in CO2-equivalents (IPCC 2007). Acidification potential was expressed as sulphur dioxide equivalents (SO2-eq) and eutrophication potential as phosphate equivalents (PO4-eq), and both were quantified according to the impact assessment method CML 2001 (Centre of Environmental Science of Leiden University (CML) 2001). The use of energy was expressed as primary energy in MJ and calculated according to CML 2001. Energy use for transport and by tractors was calculated as fossil primary energy use, since these operations use fossil energy sources, based on data taken from Ecoinvent 3.1.

The LCA was performed using the GaBi software (version 6.0, 2016, Thinkstep, Leinfelden-Echterdingen, Germany).

Inventory data

SS characteristics

The LCA was performed for mixed SS consisting of primary (mechanical), secondary (biological) and tertiary (phosphorus-precipitated) SS digested at mesophilic temperatures (25–45 °C) (Flodman 2002; Willén et al. 2016a). After digestion, the SS was assumed to be dewatered to a DM content of 29%. For the CAS treatment, it was assumed that urea was added at a rate of 1.5% of SS weight, as a source of NH3, just prior to storage. The characteristics of the SS are presented in Table 1.

Transport

It was assumed that the SS was transported directly from the wastewater treatment plant to the on-farm storage facility over a mean transport distance of 100 km, based on average transport distances in Sweden (L. Wigh, personal communication). The on-farm transportation (from storage to field) was included in the transport distance. The truck was assumed to return empty. The chemical fertilisers (TSP and AN) that were avoided by use of SS were assumed to be transported to Sweden from the Yara manufacturing site in Uusikaupunki, Finland. The transportation distance was assumed to be 500 km on water by cargo vessel and 100 km on land by truck (empty return).

Transport data were taken from Ecoinvent 3.1. All land transport was by truck with 26–28 Mg gross weight and 18.4 Mg payload capacity, using diesel fuel and with Euro 5 engines. All sea transport was conducted on a ship with a tank size of 50,000 Mg dead weight. The energy use was calculated as an average of steam turbine (5%) and diesel engine (95%) propulsion. The fuel used was heavy fuel oil.

Table 1 | Physical and chemical parameters, prior to storage, of digested and dewatered SS used in the three treatments: storage without a cover (S), storage with a cover (CS), and treatment with ammonia and storage with a cover (CAS)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>S</th>
<th>CS</th>
<th>CAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>DM</td>
<td>% of wet weight</td>
<td>29</td>
<td>29</td>
<td></td>
</tr>
<tr>
<td>Tot-C</td>
<td>% of wet weight</td>
<td>10</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>Tot-N</td>
<td>% of wet weight</td>
<td>1.2</td>
<td>1.8</td>
<td></td>
</tr>
<tr>
<td>NH4-N</td>
<td>% of wet weight</td>
<td>0.29</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Tot-P</td>
<td>% of wet weight</td>
<td>0.93</td>
<td>0.93</td>
<td></td>
</tr>
</tbody>
</table>
Nitrous oxide: For the covered treatment (CS) and the treatment with urea addition and cover (CAS), N₂O emissions data from Willén et al. (2016a) were used. Emissions data for treatment S were estimated from Willén et al. (2016a) and Flodman (2002) as described below.

The SS storage experiment with treatments S, CS and CAS described in Willén et al. (2016a) was established in autumn and the emissions decreased after approximately 70 days (around 24 November) when winter weather prevailed (i.e. the temperature fell) and then remained low for the rest of the 1-year measuring period. In contrast, the SS storage experiment (treatment S) described in Flodman (2002) was established in early summer and the N₂O emissions increased during the 116-day measuring period. To extrapolate data from that 116-day measuring period to 1 year, the rate of emissions occurring on the last four measuring occasions in that experiment (days 79–116) was assumed to continue from day 79 until the time of the year when emissions decreased according to Willén et al. (2016a), i.e. 24 November (Figure 2). Thereafter, the emissions rate was assumed to decrease in a similar way as observed from 24 November onwards by Willén et al. (2016a) for the period remaining, to cover a full year. Mean cumulative N₂O emissions from Willén et al. (2016a) and the extrapolated cumulative N₂O emissions from Flodman (2002) were then used as the default input data for treatment S in the LCA (Table 2). The emissions shown in Figures 2 and 3 in g N₂O-N and CH₄-C m⁻³, respectively, were transformed to kg N₂O and CH₄ Mg⁻¹ SS in Table 2 by dividing them by the SS density. In addition, the storage duration for the different application timings was taken into account. Indirect N₂O production
from NH$_3$ was calculated as 1% of emitted NH$_3$ during storage (IPCC 2006).

**Methane:** To get a full-year dataset of CH$_4$ emissions for treatment S, data from Willén et al. (2016a) and Flodman (2002) were combined sequentially. Flodman (2002) commenced measurements of CH$_4$ emissions on the day after establishing the storage experiment, and the emissions were then monitored for a total of 116 days. The emissions of CH$_4$ from treatments S, CS and CAS in Willén et al. (2016a) were measured for a year, but due to analytical problems the emissions during the first 100 or so days of storage could not be fully quantified and were hence omitted from the dataset used in the present study. Thus, for treatment S, up until day 116 data from Flodman (2002) were used and from day 117 onwards data from Willén et al. (2016a) were used (Figure 3). By combining these two datasets, a set covering a full year was obtained.

Until day 7 of storage, CH$_4$ emissions for treatments CS and CAS were assumed to be of the same magnitude as in treatment S, as much of the CH$_4$ emitted at the start of storage was assumed to have been produced prior to storage, when differences between the treatments had not yet developed. The emissions between days 7 and 116 for treatments CS and CAS were estimated by scaling the emission from treatment S above, according to the measured mean ratio of emissions between treatment S and each of the other treatments (CS, CAS) for the period 117–365 days in Willén et al. (2016a). For days 117–365, emissions data for CS and CAS were used, as reported by Willén et al. (2016a) just as for S (Figure 3).

**Ammonia:** For treatment S, an NH$_3$ emissions factor of 10% of incoming Tot-N, as reported for storage of semi-solid manure (Karlsson & Rodhe 2002), was used. Based on De...
Bode (1991), this factor was assumed to be reduced by 80% if the SS was covered with plastic sheeting (treatment CS and CAS).

**Cover:** The plastic cover used for CS and CAS was assumed to be made from polyethylene (0.14 kg m\(^{-2}\), 115 µm thick) and an estimated 1.56 m\(^2\) per Mg stored SS was assumed to be needed. Data on manufacture of the cover were taken from Ecoinvent 3.1.

**Storage duration:** SS is produced all year round, but the period during which application to soil is possible varies, and therefore the storage duration depends on when the SS is actually applied. The shortest storage time in all scenarios was set to 1 month due to the statutory requirement to have analytical data for the SS before application to agricultural land. For autumn application, when application of SS was assumed to be possible for 4 months, i.e. July to October (L. Wigh, personal communication), the SS needed to be stored for between 1 and 9 months. For spring application, when application was assumed to be possible for 1 month (mid-April to mid-May), the SS needed to be stored for between 1 and 12 months. The storage duration for application in autumn–spring was between 1 and 6.5 months for the part of the SS (80%) applied in autumn and between 5 and 6.5 months for the part of the SS (20%) applied the following spring.

**Land application**

Calculated direct emission rates of N\(_2\)O, CH\(_4\) and NH\(_3\) after application of SS to agricultural land are presented in Table 3. In the calculations, the emissions factor for N\(_2\)O after land application of SS was assumed to be 1% of applied Tot-N, according to IPCC (2006). Indirect N\(_2\)O-N production from NH\(_3\) was calculated as 1% of emitted NH\(_3\)-N (IPCC 2006). The emissions factor for NH\(_3\) after land application in different seasons was taken from Karlsson & Rodhe (2002) and was 25% of applied NH\(_4\)-N in autumn and 33% in spring. These values assume incorporation of SS into the soil within four hours of application. It has previously been shown in studies on land application of SS that emissions of CH\(_4\) are negligible (Ambus et al. 2001; Pitombo et al. 2015; Willén et al. 2016b), and therefore emissions of CH\(_4\) were set to zero.

The amounts of chemical N and P replaced by SS application to soil are presented in Table 4. The amount of SS applied was calculated based on its P content. The P application rate in Sweden is restricted to 110 kg P ha\(^{-1}\) during a 5-year period (Swedish EPA 2002). The amount of P in SS replacing chemical P was estimated to be 60% of Tot-P, which is towards the upper end of the 25–75% P bioavailability range in SS proposed by Foley et al. (2009). The ratio of N replacing chemical N was calculated according to Equation (1) (Delin et al. 2012):

\[
Y = 0.87 - 0.05 \times \left(\frac{C}{N}\right)
\]  

where Y is the amount of N in SS replacing chemical N, C and N are the concentrations of total carbon and total nitrogen in SS, respectively, and 0.05 and 0.87 mark the range within which a linear relationship between the C:N ratio and N availability applies. Primary energy use for the manufacture of chemical N and P was calculated based on data from Brentrup & Pallière (2014) and was 42 MJ kg\(^{-1}\) N for AN, 51 MJ kg\(^{-1}\) N for urea and 0.86 MJ kg\(^{-1}\) P for TSP. GHG emissions (mainly CO\(_2\) and N\(_2\)O) during manufacture of chemical fertilisers were also calculated based on data from Brentrup & Pallière (2014). Data on the composition of primary energy inputs for chemical N and P manufacture were taken from Davis & Haglund (1999), and conversion factors for calculating primary energy use for chemical N and P manufacture were taken from the Sustainable Energy Authority of Ireland (SEAI 2014). The tractor-drawn manure spreader for spreading SS on land was assumed to be fitted with two horizontal beaters and a control system for constant application rate. Data for tractor...
operations and fuel combustion were taken from Eco-invent 3.1.

Sensitivity analysis

Magnitude of nitrous oxide and methane emissions

The studies from which data on N2O and CH4 emissions from storage of SS were taken, and several previous studies on storage of organic fertilisers, show high variability in amounts of gaseous emissions (Flodman 2002; Chadwick et al. 1998; Willén et al. 2019). Therefore, a sensitivity analysis was performed for treatment S to test the consequences of variable (high and low) emissions of N2O and CH4 from storage on the whole storage-application system. In this sensitivity analysis, the emissions were set at 200% (high) and 25% (low) of the N2O or CH4 emissions used in the basic scenarios. The high value corresponds to the highest N2O emissions measured in Flodman (2002) and the low value is a mean of the lowest N2O emissions measured in Willén et al. (2016a) and N2O emissions reported by Börjesson & Svensson (1997). The emissions levels were varied in the same way for CH4.

Nitrate leaching

In the basic scenarios, the N availability, i.e. the N that replaced chemical fertiliser, did not take into account losses of N between application in autumn and the start of substantial crop uptake in spring. In a report commissioned by the Swedish Board of Agriculture, Aronsson & Torstensson (2004) propose a leaching factor of 0.13–0.30 for fertilised soils in Sweden, depending on the clay content of the soil. In the sensitivity analysis, 30% of the NH4-N from the SS remaining after land application was assumed to be lost by NO3 leaching in autumn-only application, and from the autumn part in the autumn-spring application. The amount of N leached affects the eutrophication potential and also the amount of chemical AN that can be avoided.

Immediate incorporation

A reduction in NH3 emissions after land application of SS can be achieved by incorporating the SS into the soil immediately after application (Karlsson & Rodhe 2002). Immediate incorporation of SS into the soil was studied in the sensitivity analysis, with the amount of applied NH4-N being emitted as NH3 after land application being reduced to 15% in this case (Karlsson & Rodhe 2002).

Carbon sequestration

When SS is applied, organic matter is added to the soil, leading to increases in C stocks and long-term C sequestration. The turnover of organic C in soil depends on many different factors, such as microbial activity, which is affected by temperature and soil moisture, for example, and therefore the prediction of the amount of C sequestration is uncertain. The C sequestration after 100 years was assumed here to be 7% of the total amount of added C, a factor previously used for digested food waste (Chiew et al. 2015) and blackwater (Spångberg et al. 2014).

RESULTS AND DISCUSSION

All nine scenarios investigated, i.e. combinations of the three different storage treatments and the three land application times, had a negative impact on the environment for all impact categories (meaning that the impact from each impact category exceeded zero), even when including the avoidance of chemical fertiliser manufacture and transport. The only exception was primary energy use for CS, where the impact was positive (less than zero) (Figures 4–7).

GWP

Out of the transport, storage and land application subsystems, storage was the main contributor to GWP (Figure 4). Similarly, Johansson et al. (2008) found that emissions of GHG from storage and land application of SS contribute most to GWP in all parts of the system in their study, regardless of whether high or low emissions factors are assumed. For the storage subsystem in the present study, N2O emissions in terms of CO2-eq were the main contributor to GWP in treatment S, while CH4 emissions were the main contributor in treatment CS. The GWP for treatment CAS was dominated by GHG from urea manufacture.

Cover

Covering the SS had adverse effects in that it reduced N2O emissions, but increased CH4 emissions (Figure 4). However, the reduction in N2O emissions was more important than the increase in CH4 emissions in terms of reducing GHG emissions, as can be seen from the 30% reduction in GWP for the storage subsystem when covered (treatment CS) compared with when left uncovered (treatment S).
Ammonia treatment

Ammonia treatment through the addition of urea had a pronounced effect in strongly reducing emissions of both N₂O and CH₄ during storage (Figure 4), and also resulted in an organic fertiliser with good agronomical properties by increasing the amount of N in the SS (Table 4). However, the substantial GHG emissions from the manufacture of urea cancelled out this beneficial effect, and for the whole system the CAS treatment contributed slightly more to GWP than the CS treatment, while both contributed far less to GWP than the S treatment.

Figure 4 | GWP divided by contributing category for the three storage treatments and three land application times. Total GWP is given at the top of each bar. S = storage of SS without a cover, CS = storage of SS with a cover, and CAS = storage of SS treated with ammonia and with a cover. AN = ammonium nitrate and TSP = triple superphosphate chemical fertiliser.

Figure 5 | Acidification potential divided by contributing category for the three storage treatments and three land application times. Total acidification potential is given at the top of each bar. S = storage of SS without a cover, CS = storage of SS with a cover and CAS = storage of SS treated with ammonia and with a cover. AN = ammonium nitrate and TSP = triple superphosphate chemical fertiliser.
Application time

The lower GWP from autumn-only application (actually application from late summer to late autumn) and autumn–spring application for the whole system compared with the spring-only application was a result of the different storage times assumed in the three scenarios (Figure 4). The short time window in which SS can be applied in spring in central Sweden (1 month) results in a longer average storage time for SS applied in spring than for SS applied in autumn, when the time window for land application is 4 months.

A reduction in storage time can be achieved by applying SS in both autumn and spring instead of only in autumn. The GWP from the storage subsystem was reduced by 12...
and 9% for S and CS, respectively, when SS was applied in both spring and autumn compared with application only in autumn. This was a direct effect of the shorter mean storage time of the SS in the former scenario. Since almost all storage-related GWP in CAS originated from urea manufacture, the difference between autumn-only and autumn-spring application was negligible for CAS.

**Acidification**

**Cover**

Most of the acidification potential originated from emissions of NH$_3$ during storage and after land application of the SS (Figure 5). The lowest acidification potential was therefore seen for the CS treatment, since the cover strongly reduced the NH$_3$ emissions during storage. Similar results from covering stored organic fertilisers have been reported by De Bode (1991), Chadwick (2005) and Sagoo et al. (2007).

**Ammonia treatment**

As in CS, the cover in CAS also reduced the NH$_3$ losses from storage. However, as urea addition increased the NH$_4^+$ content in the SS, the emissions of NH$_3$ increased somewhat during storage and strongly after land application. This resulted in CAS contributing more to the acidification potential than CS, and with spring application the impact was slightly higher for CAS than for S (Figure 5).

**Eutrophication**

As observed for acidification, most of the eutrophication potential effect originated from emissions of NH$_3$ during storage and after land application of SS. It should be noted, however, that N leaching from the field or from storage was not included in the basic scenarios. This caused the contributors to eutrophication in the LCA to display similar patterns to the contributors to acidification.

**Cover**

The eutrophication effect was reduced by covering the SS during storage, as can be seen from the difference between S and CS, since the cover reduced both N$_2$O and NH$_3$ emissions (Figure 6).

**Ammonia treatment**

The CAS treatment presented higher eutrophication potential than CS, despite both these treatments being covered (Figure 6). This was a result of higher NH$_3$ emissions after land application from the CAS with its higher content of NH$_4^+$ than from the other treated sludges. The NO$_x$ emitted during transport only marginally contributed to the eutrophication potential.

**Primary energy use**

The primary energy use for SS transport and tractor use for land application was similar for all nine scenarios (Figure 7). The saved primary energy use for avoided manufacture of chemical P fertilisers was low and did not markedly affect the total results. However, avoided manufacture of chemical N had a large impact, as it reduced the total primary energy use to almost zero for S and reversed the total primary energy use for CS to a negative value, meaning an overall saving in primary energy (Figure 7).

**Cover**

The CS treatment had slightly higher primary energy use than S due to manufacture of the cover material used, but this was more than compensated for by the additional fertilisation effect, since the cover conserved more mineral N in the SS, instead of being emitted as in S.

**Ammonia treatment**

The manufacture of urea constituted a very large part of the overall primary energy use in the CAS treatment.

**Sensitivity analysis**

**Magnitude of nitrous oxide and methane emissions**

In the sensitivity analysis, varying the N$_2$O emissions during storage in treatment S to 200% (high) and 25% (low) of the level in the basic scenarios had a pronounced effect on GWP from the whole system, yielding increases of $>70\%$ and decreases of $>50\%$ for the high and low scenarios, respectively (Table 5). Varying the CH$_4$ emissions by similar amounts had an impact of less than 20%, which in this analysis was deemed to represent no impact. The assumed increases and decreases in both gases changed the acidification potential and eutrophication potential, but these
Changes were less than 20% and hence also considered negligible.

An assumed increase and decrease in N$_2$O emissions resulted in a large increase and decrease, respectively, in the primary energy use (Table 6). This was a result of less chemical N being replaced when larger amounts of N were lost as N$_2$O, and hence more chemical N needing to be manufactured. The opposite was true when less N was lost as N$_2$O.

### Table 5 | Changes in impact categories (GPW in terms of CO$_2$-equivalents (CO$_2$-eq), acidification potential in terms of SO$_2$-eq and eutrophication potential in terms of PO$_4$-eq) in the sensitivity analysis

<table>
<thead>
<tr>
<th>Sensitivity analysis</th>
<th>Autumn application</th>
<th>Spring application</th>
<th>Autumn–spring application</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Treatment</td>
<td>CO$_2$-eq (%)</td>
<td>SO$_2$-eq (%)</td>
</tr>
<tr>
<td>Increased N$_2$O emissions from storage (200% of base value)</td>
<td>S</td>
<td>+71 n.i. n.i.</td>
<td>+71 n.i. n.i.</td>
</tr>
<tr>
<td>Decreased N$_2$O emissions from storage (25% of base value)</td>
<td>S</td>
<td>−53 n.i. n.i.</td>
<td>−53 n.i. n.i.</td>
</tr>
<tr>
<td>Increased CH$_4$ emissions from storage (200% of base value)</td>
<td>S</td>
<td>n.i. n.i. n.i.</td>
<td>n.i. n.i. n.i.</td>
</tr>
<tr>
<td>Decreased CH$_4$ emissions from storage (25% of base value)</td>
<td>S</td>
<td>n.i. n.i. n.i.</td>
<td>n.i. n.i. n.i.</td>
</tr>
<tr>
<td>NO$_3$ leaching</td>
<td>S</td>
<td>n.i. n.i. +25 n.r.</td>
<td>n.r. n.r. n.r.</td>
</tr>
<tr>
<td></td>
<td>CS</td>
<td>n.i. n.i. +40 n.r.</td>
<td>n.r. n.r. n.r.</td>
</tr>
<tr>
<td></td>
<td>CAS</td>
<td>+23 n.i. +40 n.r.</td>
<td>n.r. n.r. n.r.</td>
</tr>
<tr>
<td>Reduced NH$_3$ emissions after land application</td>
<td>S</td>
<td>n.i. n.i. n.i. n.i.</td>
<td>n.i. −27 −23 n.i. n.i. n.i</td>
</tr>
<tr>
<td></td>
<td>CS</td>
<td>n.i. −36 −29 n.i.</td>
<td>n.i. −51 −43 n.i. n.i. n.i</td>
</tr>
<tr>
<td></td>
<td>CAS</td>
<td>n.i. −34 −28 n.i.</td>
<td>n.i. −48 −43 n.i. n.i. n.i</td>
</tr>
</tbody>
</table>

SS treatments included are storage of SS without a cover (S), storage of SS with a cover (CS), and storage of SS treated with ammonia and with a cover (CAS). All treatments were applied in autumn, spring and autumn–spring. Increased impact is indicated with a plus (+) and decreased impact with a minus (−). Changes <20% were regarded as having no impact (n.i.). n.r. = not relevant.

### Table 6 | Changes in the impact category primary energy use in the sensitivity analysis

<table>
<thead>
<tr>
<th>Sensitivity analysis</th>
<th>Treatment</th>
<th>Autumn application</th>
<th>Spring application</th>
<th>Autumn–spring application</th>
</tr>
</thead>
<tbody>
<tr>
<td>Increased N$_2$O emissions from storage (200% of base value)</td>
<td>S</td>
<td>0.50 1.82</td>
<td>1.70 3.17</td>
<td>0.59 1.74</td>
</tr>
<tr>
<td>Decreased N$_2$O emissions from storage (25% of base value)</td>
<td>S</td>
<td>0.50 −0.49</td>
<td>1.70 0.60</td>
<td>0.59 −0.28</td>
</tr>
<tr>
<td>Increased CH$_4$ emissions from storage (200% of base value)</td>
<td>S</td>
<td>0.50 0.36</td>
<td>n.i.</td>
<td>0.59 0.47</td>
</tr>
<tr>
<td>Decreased CH$_4$ emissions from storage (25% of base value)</td>
<td>S</td>
<td>0.50 0.61</td>
<td>n.i.</td>
<td>n.i.</td>
</tr>
<tr>
<td>NO$_3$ leaching</td>
<td>S</td>
<td>0.50 6.06</td>
<td>n.r.</td>
<td>0.59 4.99</td>
</tr>
<tr>
<td></td>
<td>CS</td>
<td>−4.27 1.89</td>
<td>n.r.</td>
<td>−3.99 0.87</td>
</tr>
<tr>
<td></td>
<td>CAS</td>
<td>31.70 41.48</td>
<td>n.r.</td>
<td>32.15 39.75</td>
</tr>
<tr>
<td>Reduced NH$_3$ emissions after land application</td>
<td>S</td>
<td>0.50 −0.84</td>
<td>1.70 −0.71</td>
<td>0.59 −0.98</td>
</tr>
<tr>
<td></td>
<td>CS</td>
<td>−4.27 −5.77</td>
<td>−3.05 −5.92</td>
<td>−3.99 −5.83</td>
</tr>
<tr>
<td></td>
<td>CAS</td>
<td>n.i.</td>
<td>n.i.</td>
<td>n.i.</td>
</tr>
</tbody>
</table>

SS treatments included are storage of SS without a cover (S), storage of SS with a cover (CS) and storage of SS treated with ammonia and with a cover (CAS). All treatments were applied in autumn, spring and both autumn and spring. Changes <20% were regarded as having no impact (n.i.). n.r. = not relevant.
The reverse was seen for an assumed increase and decrease in emissions of CH₄, where an increase led to a reduction in primary energy use and *vice versa*, although the changes were less than 20% for some land application times (Table 6). The changes in primary energy use caused by increasing or decreasing CH₄ emissions were due to a change in the C:N ratio of the SS, which affected the amount of chemical N replaced by SS, in accordance with the equation used for calculating N availability (Equation (1)).

**Nitrate leaching**

When NO₃⁻ leaching was not accounted for, spring application had the highest total eutrophication potential. Introducing a NO₃⁻ leaching factor resulted in leaching losses contributing 20–40% of the total eutrophication potential for autumn-only and autumn–spring application and 40% for spring-only application (Table 5). The inclusion of leaching also increased the need for manufacture of chemical AN, which strongly increased the primary energy use (Table 6).

**Immediate incorporation to soil after land application**

Ammonia emissions were reduced by incorporation of SS into the soil directly after application, instead of within four hours as in the basic scenarios (Table 5). This is in line with previous findings by Misselbrook et al. (2002) and Rodhe et al. (2006). However, for treatment S with autumn-only application and autumn–spring application, the reduction was somewhat lower than 20% (indicated as no impact in Table 5). The primary energy use decreased when less NH₃ was emitted after land application, as a consequence of the smaller need for chemical AN manufacture (Table 6).

**Carbon sequestration**

Introducing a C sequestration factor corresponding to 7% of the total amount of C added to soil over 100 years resulted in a reduction in GWP of 5.5 kg CO₂-eq FU⁻¹ for treatments S and CS and 4.5 kg CO₂-eq FU⁻¹ for CAS for all land application times. The higher C sequestration for the CAS treatment was due to less C being lost through emissions of CH₄ during storage in this treatment. The inclusion of C sequestration in the system changed the impact on GWP from the land application subsystem to negative for all treatments, meaning that using SS as fertiliser decreased global warming over a 100-year perspective. Overall, the inclusion of C sequestration resulted in a reduction in GWP ranging from 14% (S applied in spring) to 56% (CAS applied in autumn and autumn plus spring) in the whole system (Figure 8).

**Application time and storage duration**

The scenarios with spring-only application showed the highest GWP, acidification potential and eutrophication potential for the total system in the basic scenarios irrespective of SS storage treatment (Figures 2–4). This higher GWP was a result of the larger emissions of N₂O and CH₄ due to the longer storage time for the spring scenarios compared with the autumn and autumn–spring scenarios. The time of year of storage, and hence differences in weather conditions, was not considered in this context. The result was therefore a consequence of the short time available for applying SS in the field during spring. The higher acidification and eutrophication potential were a consequence of the larger emissions of NH₃ when applying SS in spring compared with in autumn (Table 3). It has previously been shown that the drier soils in spring and summer lead to somewhat lower emissions of N₂O than when SS is applied in autumn when the soil contains more moisture (Willén et al. 2016b). Similarly, Pradel et al. (2013) concluded in a
model of SS application to land that N\textsubscript{2}O emissions depend strongly on the soil and prevailing weather conditions. If these circumstances had been considered in the present study, the GWP from land application would probably have been somewhat lower in spring or somewhat higher in autumn than in the basic scenarios. Furthermore, since the autumn application period starts in late summer, the simulated N\textsubscript{2}O emissions might have been slightly too high and the NH\textsubscript{3} emissions slightly too low for this application time.

The results of the LCA indicated that application only in spring was not the best option out of the scenarios studied regarding environmental impact. Moreover, due to the short time window available, spring application of all SS produced in Sweden would not be feasible without greatly increasing the number of spreaders and, consequently, the cost. Rather, a combination of application in autumn and spring would be more realistic and also seemed to be a good option, at least in terms of GWP. However, when taking into account NO\textsubscript{3} leaching and thus also less N in the SS that replaces chemical N, as suggested in the sensitivity analysis, spring application was preferable to autumn or autumn–spring application in terms of eutrophication potential and primary energy use. Spring application was also preferable to the other application times in terms of GWP for autumn–spring application for the CS treatment and for both autumn-only and autumn–spring application for the CAS treatment. In terms of acidification potential, autumn-only or autumn–spring application was still the better option, and for treatment CS with autumn–spring application and for treatment S with both autumn-only and autumn–spring application in terms of GWP as well. However, in light of the suggestions that the drier soil in spring produces lower emissions of N\textsubscript{2}O, spring application of SS seems a reasonable option.

Reduced storage time might be a good measure for mitigating the GWP from SS management, since the shorter storage time per se was not the reason for the slight increase in acidification potential observed when shifting from autumn-only application to autumn–spring application. However, since SS is produced at the same rate all year round, it is difficult to further reduce storage times. Previous studies have shown that GHG emissions during storage reduce during colder seasons (Sommer et al. 2004; Willén et al. 2016a). The data used in the present LCA did not consider the time of year and hence the weather conditions in which the SS was stored. Spring application restricted to only 1 month results in storage of SS all year round, with the oldest SS being almost 1 year old when applied to the soil. However, less SS is stored during the warm summer when the storage facility is emptied for spring application, while with autumn application the storage facility is almost full during the warm summer. In this regard, spring application is preferable. Cold storage of organic fertilisers has been shown to be an effective way of reducing GHG emissions (Sommer et al. 2004; Clemens et al. 2006). Furthermore, acidification by the addition of sulphuric acid has been proven to reduce both GHG and NH\textsubscript{3} emissions during storage (Petersen et al. 2012), but might require soil liming to counterbalance the acidity.

**Covered storage**

The results of the LCA showed that use of a cover was a good measure in terms of lowering all impact categories studied. Covering during storage is a common measure to reduce odour and losses of valuable NH\textsubscript{3} from stored organic fertilisers (De Bode 1991). Even though the CH\textsubscript{4} emissions from storage and NH\textsubscript{3} emissions after land application increased in the covered treatment compared with the non-covered treatment in the present study (Tables 2 and 3), the magnitude of all individual impact categories evaluated was lower for CS than for S (Figures 2–5).

**Ammonia treatment**

Similarly to using a cover, NH\textsubscript{3} treatment (CAS) reduced GHG emissions during storage but increased NH\textsubscript{3} emissions after land application compared with treatment S (Tables 2 and 3, Figure 4). The GWP for CAS was only slightly higher than that for CS, despite the contribution from urea manufacture in CAS. This increase in GWP was compensated for by the larger amount of chemical N avoided in CAS compared with CS (Figure 4). However, acidification potential and eutrophication were higher for CAS than for CS, and for acidification potential in spring even higher than for S as a consequence of the higher emissions of NH\textsubscript{3} due to the added urea (Figures 3 and 4).

One important aspect in choosing the SS treatment method or time for land application is achieving efficient use of plant nutrients. For example, the scenarios with CAS application in autumn are not fully applicable in reality, since there is no good reason for applying fertiliser containing much easily available N in autumn, since the crop will not take up this N until after winter. This was clearly shown by the higher NO\textsubscript{3} leaching from CAS than from the other treatments when NO\textsubscript{3} leaching was included, and also by the higher eutrophication for CAS...
with autumn and autumn–spring application compared with the other treatments in that scenario. Application of SS following CAS treatment should therefore preferably be restricted to spring, if used at all. However, it should be borne in mind that CAS storage would probably also meet any future requirement to sanitise SS before application to land (Nordin et al. 2009).

General discussion

The main contribution to GWP from recycling SS to arable land originates from gaseous emissions of GHG, either during storage of the SS or following land application. Johansson et al. (2008) showed the importance of accounting for emissions of GHG from both storage and land application when performing an LCA on SS management, with the FU being the treatment of 1 Mg DM digested SS. In LCA with and without inclusion of emissions from storage and land application, those authors found that the results changed dramatically. Their core system, consisting of transport and machine operations, contributed 48 kg CO2-eq. FU−1, while GHG emissions from SS during storage and after land application contributed 225 and 3,560 CO2-eq. FU−1 for a low (0.04 kg N2O Mg−1 wet weight) and high (1.73 kg N2O Mg−1 wet weight) N2O emissions estimate, respectively. Their high N2O emissions estimate was about three times the ‘high’ value tested in the sensitivity analysis in the present study, while their low N2O estimate was half the ‘low’ value. Similar patterns of impact were shown for acidification and eutrophication.

Improvements in the use of fuels, machinery, type of transportation and transport distances would have some impact on GWP and primary energy use. However, due to the dominant contribution to GWP from SS storage (S and CS) and urea manufacture (CAS) and to primary energy use from AN manufacture (all treatments) and urea manufacture (CAS) observed in this study, such improvements would not have a pronounced effect in these scenarios. The emissions contributing to GWP from chemical fertiliser manufacture used in the present LCA were slightly higher than those from a best available technology manufacturing system, and slightly more than half those of the mean European manufacturing system in 2006 according to Brentrup & Pallière (2014).

The results of an LCA can vary widely with the system boundaries chosen, as the magnitude of impact of different sources differs depending on the system within which they are being compared. For example, Tillman et al. (1998) found that avoided manufacture and use of chemical fertiliser did not have a great impact on environmental performance in their study on wastewater treatment and SS management, most likely due to their wide system boundary also including the wastewater treatment process. On the other hand, Lundin et al. (2000), who also studied wastewater treatment and SS management using a wide system boundary, concluded that it is important to include avoided fertilisers in the analysis. As found here, they concluded that the use of fossil resources (energy) is particularly strongly affected when avoided chemical fertiliser manufacture is included in the system. As in the present study, some scenarios in their study had such large avoided energy use that the total energy use was very small or even negative (Lundin et al. 2000).

The results of the present study show that the GHG emissions from SS storage can be substantial. Similar results have been reported for landfilled SS by Suh & Rousseaux (2002) and Houillon & Jolliet (2005), who studied different treatment strategies for wastewater sludge. Tillman et al. (1998) also concluded that GHG emissions from landfill significantly contribute to GWP, although their system boundaries were much wider than in the other studies cited. Thus, it is important to include emissions during storage in an LCA of organic fertiliser management in order to obtain a valid holistic picture.

The GHG and NH3 emissions from land application contributed more to GWP, acidification and eutrophication (when avoided emissions from chemical fertilisers were not accounted for) than GHG and NH3 emissions from storage in both the CS and CAS treatments for all application times, and therefore land application should be included in any systems analysis. This has previously been discussed by Dalemo et al. (1998), who concluded that emissions of N2O, NH3 and NO3− from SS land application may have a considerable impact on GWP and acidification potential and eutrophication.

CONCLUSIONS

In this LCA, the impacts of different storage treatments and land application times for SS were examined. The following conclusions were drawn:

- It is important to account for emissions of GHG and ammonia from storage and after land application when estimating the environmental impact of SS management.
- Shorter storage times have great potential to reduce the environmental impact from SS after application to land,
The magnitude of N$_2$O emissions during storage of SS

- **Systems with autumn application are preferable to systems with spring application for all impact categories if nitrate leaching is excluded.** When nitrate leaching is included, spring application is preferable in terms of eutrophication potential and primary energy use and, for covered storage and ammonia-treated sludge covered during storage, in terms of GWP. A combination of autumn and spring application is preferable to autumn-only application with/without nitrate leaching included, except in the case of ammonia-treated, covered sludge with nitrate leaching excluded.

- **Covered storage in combination with land application has a lower impact in terms of GWP, acidification potential, eutrophication potential and primary energy use than non-covered storage.**

- **Ammonia treatment and covering of sludge during storage is a better option than no treatment and no cover in terms of GWP, acidification potential and eutrophication potential.** While this treatment has very high primary energy use due to the energy-intensive manufacture of urea used for ammonia treatment, the ammonia sanitises the sludge during storage.

- **The magnitude of N$_2$O emissions during storage of SS has a great influence on GWP and primary energy use in the whole system.**

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