Technical University of Denmark

Environmental Assessment of Sewage Sludge Management – Focusing on Sludge Treatment Reed Bed Systems

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Publication date:
2017

Document Version
Publisher's PDF, also known as Version of record

Citation (APA):
Larsen, J. D., Scheutz, C., & Nielsen, S. (2017). Environmental Assessment of Sewage Sludge Management – Focusing on Sludge Treatment Reed Bed Systems. Kgs. Lyngby: Department of Environmental Engineering, Technical University of Denmark (DTU).

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Julie Dam Larsen

PhD Thesis
June 2017

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DTU Environment
Department of Environmental Engineering
Technical University

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This PhD project followed the Industrial PhD Programme offered by Innovation Fund Denmark (Innovationsfonden) and was conducted as a collaboration between The Technical University of Denmark (DTU) and Orbicon A/S.

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Preface

The research for this PhD thesis was carried out at the Department of Environmental Engineering of the Technical University of Denmark (DTU) under the supervision of Professor Charlotte Scheutz and at the Danish environmental consultancy Orbicon A/S under the supervision of Senior Consultant Steen Nielsen. The project followed the Industrial PhD programme offered by Innovation Fund Denmark (www.innovationsfonden.dk).

The thesis is organized in two parts: the first part puts into context the findings of the PhD in an introductive review; the second part consists of the papers listed below.

I Larsen, J. D., Nielsen, S., Scheutz, C. 2017. Greenhouse gas emissions from the mineralisation process in a Sludge Treatment Reed Bed system: seasonal variation and environmental impact. *Ecological Engineering. In press.*

II Larsen, J. D., Nielsen, S., Scheutz, C. 2017. Gas composition of sludge residue profiles in a Sludge Treatment Reed Bed between loadings. *Water Science and Technology. Accepted for publication, May 2017.*

III Larsen, J. D., Nielsen, S., Scheutz, C. 2017. Assessment of Danish Sludge Treatment Reed Bed system and a stockpile area, using substance flow analysis. *Water Science and Technology. Accepted for publication, May 2017*

IV Gómez-Muñoz, B., Larsen, J. D., Bekiaris, G., Scheutz, C., Bruun, S., Nielsen, S., Jensen, L.S. Nitrogen mineralisation and greenhouse gas emission from the soil application of sludge from reed bed mineralisation systems. *Journal of Environmental Management. Under revision, May 2017.*

V Larsen, J. D., ten Hoeve, M., Nielsen, S., Scheutz, C. 2017. Life cycle assessment comparing the treatment of surplus activated sludge in a sludge treatment reed bed system with mechanical treatment on centrifuge Submitted to *Journal of Cleaner Production, May 2017.*

VI Nielsen, S., Larsen, J. D. 2017. Operational technology, economic and environmental performance of Sludge Treatment Reed Bed systems–based on 28 years of experience. *Water Science and Technology*. DOI: 10.2166/wst.2016.295

In this online version of the thesis, papers I-VI are not included, but can be obtained from electronic article databases, e.g. via www.orbit.dtu.dk or on request from DTU Environment, Technical University of Denmark, Miljoevej, Building 113, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk.

Acknowledgements

One of the most striking things I learned during the last years is that carrying out a PhD project and scientific research in general, is a long, versatile process with many facets. Behind every single page of a thesis or a scientific paper are hundreds of thoughts shared, meetings held, opinions discussed, decisions made (and changed), hours spend on rainy field trips and in buzzing laboratories, cups of coffee consumed in late evenings and just many people, who all played a role in order to make everything happen. In short, I am the author on this thesis, but definitely not the only person behind the project, so I would like to thank:

- My supervisors Steen Nielsen (Orbicon A/S) and Charlotte Scheutz (DTU Environment) for guiding me through this project. It has been a very interesting experience to work in the interface between a university and a company, and I am very grateful that I got this opportunity. Thank you for supervising me, encouraging me, educating me and challenging me, you taught me a lot.
- The staffs at the wastewater treatment plants in Helsinge (Grib Vand), Himmark (Sønderborg Forsyning) and Stenlille (Sorø Forsyning) for letting us use their facilities as experimental sites and for assisting in organising and conducting the various activities. Thanks to Grib Vand for providing operational data for the project.
- The laboratory staff at DTU, especially Susanne Kruse and Sinh Hy Nguyen, for guiding me through various laboratory work, and Bent Skov for assisting in the making of field equipment.
- The department of Plant and Soil Sciences at the University of Copenhagen (KU) for providing laboratory facilities and assistance related to a specific part of the research.
- My colleagues at Orbicon A/S for providing a very nice and friendly work atmosphere. Thanks to Martin Støvring, Maria Laugen and Esben Bruun for helping me gathering information, data etc. on different issues related to the project and for being good colleagues. Special thanks goes to Esben, with whom I shared office, for cheering me, supporting me and discussing world affairs with me, especially those happening in Essos and Westeros. And thank you, Anders Christensen, for supporting me and helping me keeping the big overview in many ways, your help really made a difference.
- My colleagues at DTU, also for providing a very nice, friendly and supporting work atmosphere. Special thanks go to Nynne Nørup, my office mate, for support, candy and friendship, inside as well as outside the office walls.

When I signed up for this PhD project, I moved from Aarhus to Copenhagen. Now, four years later, I have grown fond of living in the capital, and will presumably stay here for a while. My friends from my years as a biology student in Aarhus still live “at the other side”. Nevertheless, we are still in touch, weekly through red-hot phone signals and by Facebook, but certainly also in real life, however not so often. When I moved from Aarhus I was afraid that we would lose contact; we did not. Maria, Randi, Anne, Signe, Lea, Kirsten, Bo, thank you so much for being there and being who you are. And thank you so much to my family for being who they are. The most striking thing I learned during the last years is that friends and family are far more valuable than any grade or title.

Summary

Sewage sludge is generated from the treatment of domestic wastewaters at wastewater treatment plants. Since the implementation of stricter requirements for wastewater treatment in the European Union in 2005, the amount of sludge produced has increased, creating the demand for more effective treatment and recycling.

In Denmark, the application of sludge on agricultural land is an often-used recycling strategy, as it returns nutrients and microelements to the soil, which can substitute for commercial fertilisers. Conventionally, sludge produced in Denmark is dewatered with mechanical devices; however, in the late 1980s, sludge treatment reed bed (STRB) systems were introduced in Denmark and in 2016, more than 100 STRB systems were operating in the country. Sludge treatment in STRB systems is often considered more environmentally friendly compared to mechanical sludge treatment technologies, albeit only a few life cycle assessments (LCAs) comparing the environmental performances of sludge treatment technologies include STRB systems. Furthermore, as data on the STRB system technology suitable for LCA are scarce, the results of these LCAs are unreliable.

The project aimed at generating data on the STRB system technology that would be useable for LCA. Based on identified knowledge gaps, research focused on three areas; quantification of gas emissions directly related to treatment, establishment of substance flows through the technology and the fate of carbon and nitrogen-based compounds in treated sludge when applied to the land. The overall goal of the project was to perform an LCA comparing the environmental performance of the STRB system technology with a conventional technology based on mechanical dewatering of sludge on a decanter centrifuge and subsequent storage. Geographically, the project focused on Denmark, and was carried out as a collaborative effort between the Technical University of Denmark (DTU) and the Danish environmental consultancy Orbicon A/S. The outcome of the project was a dataset on the STRB system technology usable for LCA, and an LCA comparing the environmental profiles of the STRB system technology and a mechanical treatment technology, constituting a basis for decision-making in relation to choice of technology.

A major part of the project involved performance of fieldwork and laboratory work. Data were collected at three Danish, well-operated STRB systems; furthermore, data required to represent the mechanical treatment technology were collected alongside data on STRB systems. Most of the data collection was undertaken at a wastewater treatment plant housing both technologies, thereby making it possible to make the two datasets as comparable as possible.

Fourteen environmental impact categories were included in the LCA, and the environmental loadings and saving provided by the sludge treatment technologies normalised to represent the treatment of 1000 kg wet weight of sludge. The life cycle inventory and the choices underlying the life cycle impact assessment were based on international acknowledged standards and recommendations. An attributional LCA approach was chosen, and the loadings and savings for all impact categories were normalised to people equivalents (PE) (the annual loadings and savings provided by one average person). Three sludge treatment scenarios were defined: 1) mechanical treatment on centrifuge, followed by storage and finally land application, 2) treatment in an STRB system and finally land application (S-STRB), and 3) treatment in an STRB system, followed by post-treatment on a stockpile area (SPA) and finally application (S-SPA).

The project succeeded in generating data on STRB systems, which could form the basis for a LCA, and comparable data related to mechanical sludge treatment. The results of the LCA revealed that STRB systems performed comparable to or better than mechanical treatment. The two scenarios based on the STRB system technology (S-STRB and S-SPA) performed comparable which only minor differences.

According to toxic impact categories, which for both technologies were mainly impacted by metals contained by treated sludge applied on land, the three scenarios performed comparable. Indeed, the substance flow analyses revealed that the metals held by sludge subjected to treatment for all scenarios were accumulated in the final sludge product. For all scenarios, the net-loadings for the impact categories Human Toxicity – Non-Carcinogenic and Ecotoxicity corresponded to $2.0 \cdot 10^{-2}$ PE, and for Human Toxicity – Carcinogenic to $5.0 \cdot 10^{-4}$ PE.

Emission rates of CO_2, CH_4 and N_2O related to biological processes in sludge subjected to treatment in STRB systems were measured during all four seasons of the year. The results revealed that seasonal variations were considerable, and should be taken into account when calculating annual, average emission rates. The emission rate of CO_2 measured from external storage of mechanically treated sludge was much lower compared to those measured for STRB systems, reflecting a lower microbial activity in the mechanical dewatered sludge. As the emission rates of the potent greenhouse gasses CH_4 and N_2O were larger for mechanical dewatered sludge, the net environmental loadings provided to the impact category Climate Change by this technology (S-CEN) and the STRB system technology (S-STRB and S-SPA) ended up being equally sized ($9.0 \cdot 10^{-4}$ PE), despite of higher biological activity in the STRB systems.

As a consequence of the lower microbial activity in mechanically treated sludge, the concentration of carbon and nitrogen-based compounds in the final sludge product produced by this treatment technology was higher compared to the final sludge product produced by treatment in STRB systems. Hence, the loadings affecting impact categories related to eutrophication and acidification were higher for the mechanical treatment technology, especially in relation to the category Marine Eutrophication, the net-loadings to this category being $8.0 \cdot 10^{-4}$ PE for mechanical treatment (S-CEN) and $3.0 \cdot 10^{-4}$ PE for STRB systems (S-STRB and S-SPA).

The STRB system technology consumed fewer abiotic resources, due mainly to the fact that the mechanical treatment process requires an input of polymer coagulant, while a STRB system does not require this contribution. Furthermore, as mechanically treated sludge often have a stronger odour compared to sludge treated in STRB systems, the latter is often claimed by the local land application sites, while mechanically treated sludge often must be transported longer distances to land application sites willing to apply it. Hence, the STRB system technology required a lower input of fuel for transportation.

In the future, it would be relevant to use the obtained data on STRB systems to compare the technology with other sludge treatment technologies commonly used. Furthermore, it would be relevant to generate a comparable dataset on representing the performance of the technology in other climate zones, and to expand the data set with more data related to economics, making it possible to make more detailed economical assessments.

Dansk sammenfatning

Spildevandsslam er restproduktet dannet ved rensning af spildevand fra husstande. Siden implementeringen af strengere krav til rensning af spildevand i den Europæiske Union i 2005 er produktionen af slam steget markant, hvilket øger efterspørgslen for mere effektiv behandling og genanvendelse af slam.

I Danmark er en af de mest anvendte genanvendelsesstrategier for spildevandsslam udbringning på landbrugsjord, da dette giver mulighed for at recirkulere næringsstoffer og mikroelementer, og derved erstatter brugen af handelsgødning. I Danmark behandles spildevandsslam konventionelt via mekanisk afvanding. I slutningen af 1980'erne blev der imidlertid indført en alternativ slambehandlingsmetode, biologiske slamanlæg (BSA). I 2016 var der i Danmark mere end 100 BSA i drift. Behandling af slam i BSA betragtes ofte som mere miljøvenlig i forhold til konventionelle slambehandlingsmetoder. Der er imidlertid kun udført få undersøgelser med formål at vurdere de miljømæssige effekter ved brug af BSA ift. andre slambehandlingsmetoder. Grundet et sparsomt datagrundlag for BSA er resultaterne af allerede udførte miljøvurderinger desuden behæftet med væsentlig usikkerhed.

Formålet med projektet var at udføre en miljøvurdering af behandling af spildevandsslam i BSA, og at sammenligne denne med mekanisk behandling på centrifuge og efterfølgende oplagring. Projektet fulgte Erhvervs Ph.d. Programmet, udbudt af Innovationsfonden, og foregik som et samarbejde mellem Danmarks Tekniske Universitet (DTU) og den danske miljøingeniørvirksomhed Orbicon A/S. Resultatet af projektet var et metodespecifikt datasæt for BSA til brug i livscyklusvurderinger, og en livscyklusvurdering af BSA og mekanisk behandling, baseret på danske forhold.

For at opnå pålidelige resultater var målet at generere nye data repræsentative for behandling af slam i BSA. Tre fokusområder blev valgt: Kvantificering af biologiske gasemissioner fra selve behandlingsprocessen i BSA, kortlægning af massestrømme gennem behandlingsprocessen og en undersøgelse af dynamikken i omdannelse og udvaskning af kulstof- og nitrogenforbindelser i det færdigbehandlede slamprodukt i forbindelse med udbringning på landbrugsjord. For at gøre vurderingen af de to behandlingsmetoder så præcis som muligt, blev der for de samme fokusområder også indsamlet data repræsentative for mekanisk behandling af slam.

Felt- og laboratoriearbejde udgjorde en væsentlig del af arbejdsprocessen. Data blev indsamlet fra tre danske BSA kendt for at være veldrevne og for at levere et færdigt slamprodukt af god kvalitet. Endvidere blev data, repræsentative for mekanisk behandling af slam og for de samme fokusområder, også indsamlet. For at gøre data

for de to behandlingsteknologier så sammenlignelige som muligt, blev størstedelen af data indsamlet på et renseanlæg, som anvender både BSA og mekanisk afvanding på centrifuge.

Livscyklusvurderingen inkluderede 14 miljøpåvirkningskategorier. De miljømæssige bidrag blev normaliserede til at repræsentere de miljømæssige påvirkninger som følge af behandling af 1000 kg slam (vådvægt). Livscyklusvurdering fulgte de internationale standarder for livscyklusvurderingsprincippet. En attributionel tilgang til vurderingen blev valgt, hvilket betyder at sammenligningen tager udgangspunkt i teknologiernes aktuelle formåen. For alle påvirkningskategorier blev bidragene normaliserede til personækvivalenter (PE), hvor én PE repræsenterer det årlige bidrag produceret af én gennemsnitlig person. Tre scenarier for slambehandling blev opstillet: 1) mekanisk behandling, efterfulgt af oplagring udbringning på landbrugsjord (S-CEN), 2) behandling i BSA efterfulgt af udbringning på landbrugsjord (S-STRB) og 2) behandling i BSA efterfulgt af efterbehandling på omlasteplads og udbringning på landbrugsjord (S-SPA).

Målsætningen om at producere et datasæt for BSA brugbart i livscyklusvurderinger, samt et datasæt, repræsentativt for mekanisk behandlet slam, blev nået. Biologiske slamanlæg viste sig at klare sig tilsvarende eller bedre end mekanisk slambehandling. Miljøpåvirkningen forårsaget af de to scenarier baseret på BSA (S-STRB og S-SPA) var stort set ens.

I forhold til toksikologiske effekter var miljøpåvirkningen den samme for alle tre scenarier, svarende til $2.0 \cdot 10^{-2}$ PE for påvirkningskategorierne Human Toksicitet – Ikke-kræftfremkaldende stoffer og Økotoksicitet, og $5.0 \cdot 10^{-4}$ PE for kategorien Human Toksicitet – Kræftfremkaldende stoffer. Toksikologiske effekter blev primært forårsaget af metaller, hvilke for alle tre scenarier blev opkoncentreret i det færdigbehandlede slam, og derved ultimativt udbragt på landbrugsjord.

Emissionsrater for CO_2, CH_4 og N_2O fra biologiske processer i slam under behandling i BSA, blev målt for alle fire årstider. Resultaterne viste, at årstidsvariationer giver anledning til væsentlige udsving i emissionerne af de nævnte gasarter, og derfor bør inddrages, når gennemsnitlige årsrater beregnes. For mekanisk behandlet og efterfølgende oplagret slam var emissionsraten af CO_2 meget lavere end for BSA, hvilket afspejler mindre biologisk aktivitet i mekanisk behandlet slam. Den procentvise andel af kulstof og nitrogen omdannet til de potente klimagasser CH_4 og N_2O var derimod lavere for BSA, hvilket betød at de miljømæssige bidrag til påvirkningskategorien Klimaforandringer var lige store for BSA (S-STRB og S-SPA) og mekanisk behandling (S-CEN), begge $9.0 \cdot 10^{-4}$ PE, på trods af højere biologisk aktivitet i BSA.

Som følge af den lavere biologiske aktivitet i oplagret, mekanisk behandlet slam viste massestrømsanalysen at koncentrationerne af kulstof- og nitrogenforbindelser i det færdigbehandlede slamprodukt, produceret af denne teknologi, var højere end i det færdigbehandlede slamprodukt fra BSA. Derfor var bidragene til påvirknings-kategorier relateret til eutrofiering og forsuring højere fra mekanisk behandlet slam, især for kategorien Marin Eutrofiering, hvor det samlede bidrag fra mekanisk behandlet slam (S-CEN) udgjorde $8.0 \cdot 10^{-4}$ PE, mens det for slam, behandlet i BSA (S-STRB og S-SPA), udgjorde $3.0 \cdot 10^{-4}$ PE.

Behandlingsprocessen knyttet til BSA havde et lavere forbrug af abiotiske ressourcer, hovedsageligt på grund af at den mekaniske behandlingsproces kræver et input af polymermasse, hvilket BSA ikke gør. Desuden giver mekanisk behandlet slam ofte anledning til lugtgener, mens slam, behandlet i et velfungerende BSA, er uden lugt. Dette betyder, at slam behandlet i BSA typisk hurtigt bliver afsat til landbrug i lokalområdet, mens mekanisk afvandet slam må transporteres over længere afstande til landbrug, der er villige til modtage det, hvilket resulterer i et højere forbrug af brændstof i forbindelse med transport.

I fremtiden vil det være relevant at bruge de nygenererede data for BSA til at udføre livscyklusvurderinger, der sammenligner teknologien med andre ofte anvendte slambehandlingsmetoder. Det vil desuden være relevant at skabe lignende datasæt for BSA beliggende i andre klimazoner, og at udvide datasættende med detaljer om de økonomiske aspekter af behandlingsprocessen.

Table of Contents

Abbreviations

DTU: Technical University of Denmark

DW: Dry Weight

FU: Functional Unit

GWP: Global Warming Potential

LCA: Life Cycle Assessment

LCIA: Life Cycle Impact Assessment

SAS: Surplus Activated Sludge

SPA: Stockpile Area

STRB: Sludge Treatment Reed Bed

WW: Wet Weight

WWTP: Waste Water Treatment Plant

1 Background

1.1 Treatment of sewage sludge in Denmark

Sewage sludge is a residual waste product generated from the treatment of domestic wastewaters at wastewater treatment plants (WWTPs). Since the implementation of the Urban Wastewater Treatment Directive (UWWTD) (91/271/EEC) in 2005, which requires more extensive and effective treatment of wastewater, sludge production has increased in the European Union by 50% (Fytili & Zabaniotou 2008). The amounts and composition of the sludge produced from a WWTP depend on the properties of the influent wastewater and the wastewater treatment processes employed. Several alternatives are available for recycling sludge, such as landfilling, incineration, industrial processes, anaerobic biological treatment and energy recovery, or its use as a fertiliser on agricultural land (Jensen and Jepsen 2005; Rovira et al. 2011). In Denmark, recycling sludge by spreading it on agricultural land is often the preferred recycling technology, because it returns nutrients and microelements to the soil, which can then replace chemical fertilisers (Oleszkiewicz and Mavinic 2002; Council of the European Union 2009).

In general, sewage sludge has low dry solid (DS) content (0.4-3%) and high contents of organic matter, nutrients, metals, xenobiotics categories and pathogens (De Maeseneer 1997; Singh and Agrawal 2008; Annabi et al. 2011). Due to the high water content of sludge, dewatering is needed to reduce its volume, thereby making it easier to handle and reducing costs involved in transportation and disposal. The contents of metals, xenobiotics and pathogens must be reduced to avoid environmental hazards such as water resource contamination, the bioaccumulation of heavy metals or xenobiotics categories or epidemic outbreaks. Furthermore, in cases where the treated sludge is applied to the land, the content of organic matter and nutrients must be stabilised, in order to prevent eutrophication of the surrounding environment.

Conventionally, sludge produced in Denmark is dewatered by using mechanical devices such as centrifuges, screw presses or filter presses (Jensen & Jepsen 2005). Before mechanical dewatering, the sludge often needs to be pre-conditioned by pre-thickening and adding polymer coagulants. After dewatering, the dewatered sludge can be processed further according to the chosen strategy for final disposal. If the dewatered sludge is going to be applied to the land, it must be stored until spring or autumn, when agricultural land fertilisation is undertaken. Some larger WWTPs have internal facilities to store dewatered sludge for short periods (1-2 weeks), before it must be transferred to a more spacious, external storage facility;

however, WWTPs rarely have the capacity to store the entire production of dewatered sludge until the time for land application. Furthermore, dewatering and storage procedures often involve considerable expense for the plant operator (Nielsen 2015). Therefore, due to a lack of storage capacity and resources, many WWTPs do not have their own dewatering or storage facilities, and so the sludge from such plants is transferred daily to larger WWTPs. However, such arrangements can also involve extensive costs; indeed, the sludge treatment activities can constitute 20-60% of the total expenses required to run an entire WWTP (Wei *et al.* 2003; Sperling & Goncalves 2007).

While stored, the dewatered sludge is often not subjected to any specific treatment, and the content of organic matter and nutrients is reduced due to mineralisation processes carried out by microorganisms; however, these processes are commonly not facilitated or optimised (Miljø- og Fødevareministeriet 2000).

In the late 1980s, sludge treatment reed bed (STRB) systems, an alternative, holistic sludge management technology combining dewatering, mineralisation and sludge storage, was introduced in Denmark (Nielsen 2003). By 2016, more than 100 STRB systems were operating in the country. The STRB system treatment method is also employed in other European countries, e.g. France (Vincent *et al.* 2011), Italy (Peruzzi *et al.* 2013), Spain (Uggetti *et al.* 2009) and United Kingdom (Nielsen & Cooper 2011). An STRB system consists of a number of beds in which the sludge being subjected to treatment accumulates over several years. Sludge accumulated in STRB systems is referred to as “sludge residue”, which is dewatered due to gravitational drainage, evaporation and water uptake by reeds growing on top of it. In addition to being dewatered, organic matter contained by the sludge is reduced, due to mineralisation by microorganisms (Nielsen 2003). The technology has also proved effective in terms of reducing xenobiotic content (Miljø- og Fødevareministeriet 2000; Nielsen 2005b). Eventually, the beds are emptied and the treated sludge applied to agricultural land.

Sludge treatment reed bed systems are often considered more environmentally friendly compared to conventional treatment technologies (Uggetti *et al.* 2010; Nielsen & Bruun 2015). However, only a few studies comparing the environmental performances of sludge treatment technologies include STRB systems. The life cycle assessment (LCA) approach, which was first developed in the 1960s (Guinee *et al.* 2010), is standardised by the International Organization for Standardization (ISO 14040 and 14044) and is used widely to evaluate the environmental performance of services or products in all community sectors, as well as in sewage sludge management. The LCA approach considers every step involved in the service or the production of a product, calculates the environmental impacts caused by each step in the process and finally gives an overview of the total environmental impact caused by the process as a whole. The approach is a useful decision-making tool, as it makes it possible to compare the performance of different services or products serving the same purpose.

Yoshida *et al.* (2013) reviewed 35 published studies evaluating sewage sludge treatment technologies by using the LCA. Out of these studies, only one, by Uggetti *et al.* (2011), included STRB systems. In addition to Uggetti *et al.* (2011), the Danish Environmental Ministry has performed an LCA study that included STRB technology (Kirkeby *et al.* 2005; Kirkeby *et al.* 2013). In these studies, STRB systems performed well compared to most other methods. However, data relating to STRB system technology suitable for LCA is scarce; therefore, the results were considered uncertain. Based on these examples, the present study aimed at generating new data on STRB systems, specifically suited for use in LCAs.

1.2 Goal and Scope of the Project

The overall goal of the project was to perform an LCA comparing the environmental impacts of treatment of sludge in STRB systems with a mechanical technology, namely sludge centrifuging, and subsequent storage. Geographically, the project focused on Denmark and aimed at producing new data on specific impact categories covering the most important knowledge gaps in relation to STRB systems. Fieldwork, laboratory work and data processing related to the generation of new data were planned and performed with LCA usability in mind. LCA modelling was performed according to the guidelines stated in ISO 14040 and 14044 and by using EASETECH, a software package developed specifically to perform LCAs on waste products. The project was carried out as collaboratively between The Technical University of Denmark (DTU) and the Danish environmental consultancy Orbicon A/S. The project followed the Industrial PhD programme, founded by Innovation Fund Denmark (Innovationsfonden, www.innovationsfonden.dk).

Knowledge gaps

Data related to the daily operation of STRB systems (electricity consumption, transport requirements) are available from the WWTPs and utilities implementing the technology. The production of electricity and the combustion of fossil fuels from transport vehicles cause emissions of carbon dioxide (CO_2), nitrogen oxides (NO_x), sulphur oxides (SO_x) and other substances, causing a range of environmental impacts. However, the LCA study by Kirkeby et al. (2013) suggested that a large proportion of the environmental emissions caused by

STRB systems are related to steps in the treatment process not requiring energy inputs, namely gas emissions caused by the mineralisation of organic matter in treated sludge.

Only a few studies have addressed gas emissions from STRB systems: Uggetti *et al.* (2012b) measured emissions of nitrous oxide (N_2O) and methane (CH_4) from an STRB system in Spain; however, this study only covered the summer season and did not include emissions of CO_2. Carbon dioxide originating from wastewater treatment or sludge treatment processes is considered short-cycled carbon and thereby climate-neutral (IPCC 2007); hence, CO_2 caused by sludge mineralisation residue does not count in the total global warming potential (GWP) of the treatment process. However, when identifying the flow of carbon through the treatment process, which is of importance when doing an LCA, carbon emitted into the air is of concern. As gas emissions in natural, reed-dominated habitats vary considerably across seasons (Søvik *et al.* 2006; Søvik & Kløve 2007), it is assumed that gas emissions from STRB systems are also affected in this regard. Furthermore, Denmark and Spain are in different climate zones, and so because microbial activity is influenced by temperature and moisture, any comparison of microbial activity carried out under different climate conditions should be undertaken with a certain degree of caution.

Cui *et al.* (2015) measured gas emissions from a pilot-scale STRB system in northern China (Dalian). This study recorded emissions of CO_2 and CH_4 over three seasons, but not emissions of N_2O, which is a potent greenhouse gas having a GWP of 265 CO_2 equivalents (excl. carbon feedbacks) (IPCC 2014), which makes it crucial to include when calculating total impact on climate change. Olsson *et al.* (2014) recorded gas emissions from a Danish STRB system; however, this study did not include N_2O either, and it only covered one season.

Knowledge on the flow of substances through the treatment process is crucial when calculating the amount of specific substances accumulated in the final sludge product that eventually will be applied to the land. Various studies have addressed changes in concentrations of substances in sludge residue as a function of treatment time in STRB systems (Pempkowiak & Obarska-Pempkowiak 2002; Peruzzi *et al.* 2013; Nielsen *et al.* 2014), but research on the flow of substances through the treatment process is not reported in the scientific literature.

When biosolids such as treated sludge are applied to the land as fertiliser, carbon (C), nitrogen (N), phosphorous (P), potassium (K), metals and other substances contained in the biosolid enter the environment. A share of these substances is taken up by the crop; however, another portion will end up in soil, groundwater, surface water or the ocean, or be emitted as gaseous compounds into the air. Therefore, the fate of substances related to land application is of great importance for an LCA covering sludge treatment technologies. Yoshida *et al.* (2015) investigated the fate of N and C in sludge treated in different scenarios in relation to land application. Even though the study included sludge from an STRB system, the data produced were not processed in a way that could be used in an LCA.

Recently, an additional procedure has been added to STRB systems. Common practice is to excavate sludge residue from an STRB system during late summer/autumn, immediately before transporting and subsequent application to agricultural land. However, in recent years, a new procedure related to emptying has been implemented for some STRB systems, whereby the beds are emptied in spring, subject to post-treatment on a stockpile area (SPA) during the summer months until application in autumn. As this procedure has only been developed recently, data on gas emissions, substance flows or the fate of substances in relation to land application are not available in the literature. As the LCA should reflect the state of the art of STRB systems, these data should also be generated.

One of the most commonly used mechanical devices for dewatering sludge is the decanter centrifuge (Jensen & Jepsen 2005). Therefore, a scenario representing a conventional sludge treatment technology was based on dewatering by centrifuge, followed by storage until the next season for land application.

When comparing different treatment technologies with an LCA, it is crucial that all treatment scenarios are treating the same type of sludge. To be able to obtain data fulfilling this requirement, a WWTP with both an STRB system and a centrifuge was chosen as main reference site.

Research areas

Based on these considerations, three research areas were defined:

Research area 1: Quantification of biological gas emissions from sludge treatment and storage

Surface emissions of CO_2, CH_4 and N_2O, released from mineralised sludge subjected to treatment in an STRB system and sludge stored after dewatering on a centrifuge, were measured by use of static flux chambers. To investigate the significance of seasonal variations in gas emissions from STRB systems, discharges from sludge were measured during spring, summer, autumn and winter. In addition, any changes in gas composition in the pore space of sludge subjected to treatment in an STRB system in relation to the dewatering process were investigated.

Research area 2: Substance flow analysis of sludge treatment scenarios

Based on samples of sludge, reject water and treated sludge collected from STRB systems, SPAs and a centrifuge, substance flows for the different treatment technologies were established.

Research area 3: Emissions from treated sludge when applied to the land

Samples of sludge treated in STRB systems, SPAs and with a centrifuge were incubated under conditions simulating land application on Danish agricultural land. During the incubation period, emissions and the accumulation of relevant substances were recorded. The obtained data were processed to express environmental emissions caused by treated sludge applied to the land over a 100-year time span.

Structure of the thesis

Data generated for the three research areas were collected at three reference STRBs located at three Danish WWTPs. Figure 1 provides an overview of the data generation process.

The following chapters are as follows: STRB systems are described in Chapter 2, “Sludge treatment reed bed systems”, after which the reference WWTPs and the methodology behind the data generation and the LCA are described in Chapter 3 “Methodology”. In Chapter 4, “Results”, the main results related to the three research areas are presented, followed by a presentation and interpretation of the LCA results. In Chapter 5, “Conclusions”, the main conclusions of the project are summarised, and finally, in Chapter 5, “Further Research”, thoughts on future perspectives and further investigation are discussed briefly.

The project followed the Industrial PhD programme offered by Innovation Fund Denmark (www.innovationsfonden.dk). An Industrial PhD project is carried out collectively between a university, in this case DTU, and a company.

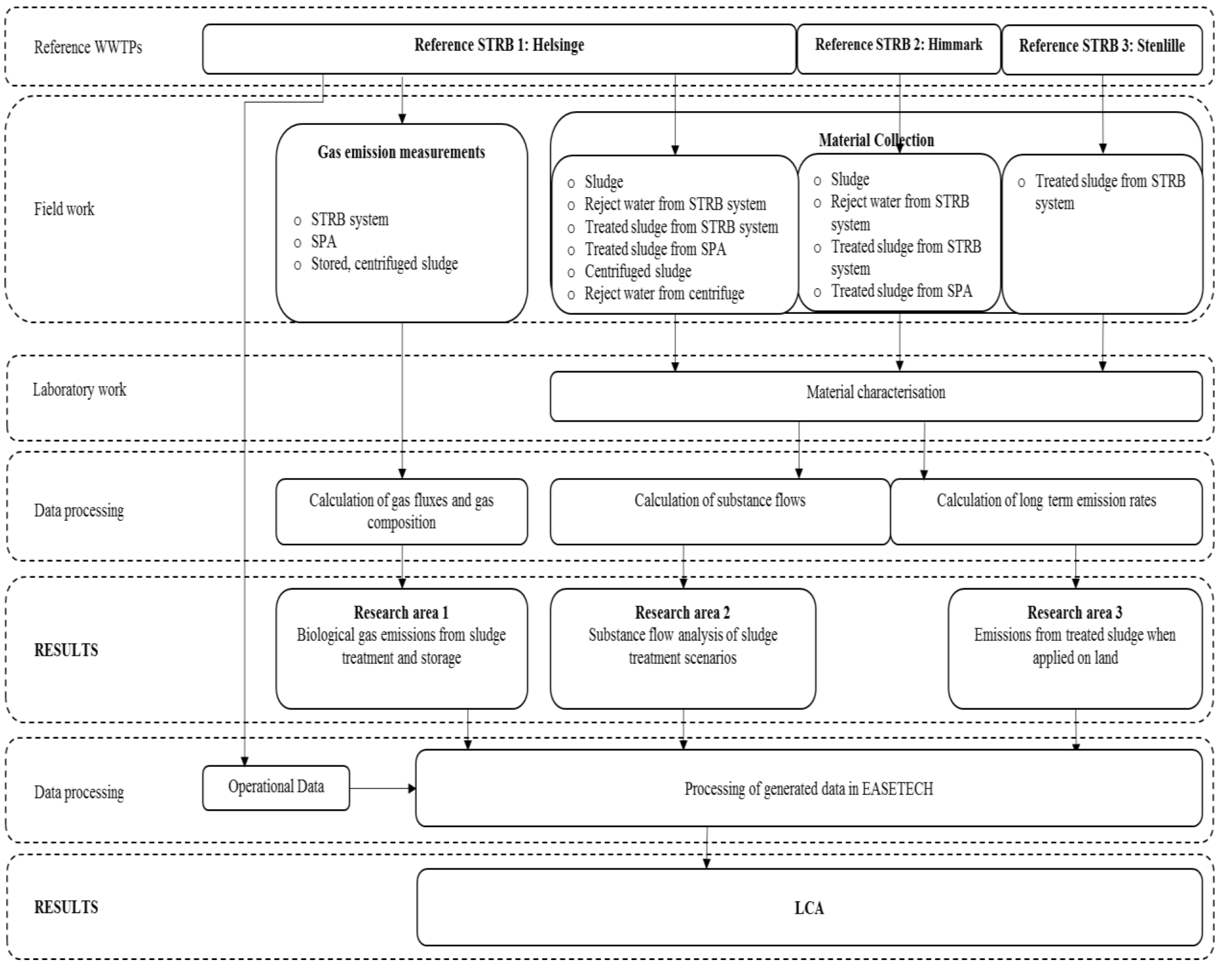

Figure 1. Overview of the data generation process throughout the project.

The Industrial PhD programme implies that the PhD candidate is employed by the company and enrolled in the PhD programme provided by the university. The collaborating company was Orbicon A/S, a Danish environmental consultancy.

2 Sludge treatment reed bed systems

This chapter provides an overview of the design and operational procedures involved in operating an STRB system.

2.1 Design and operation

Sludge treatment reed bed systems are commonly constructed close to a WWTP, from which they receive sludge (Figure 2a). An STRB system consists of a number of beds, commonly eight or ten, though they come in many sizes and various numbers of beds, depending on the capacity required for treatment (Nielsen & Willoughby 2005).

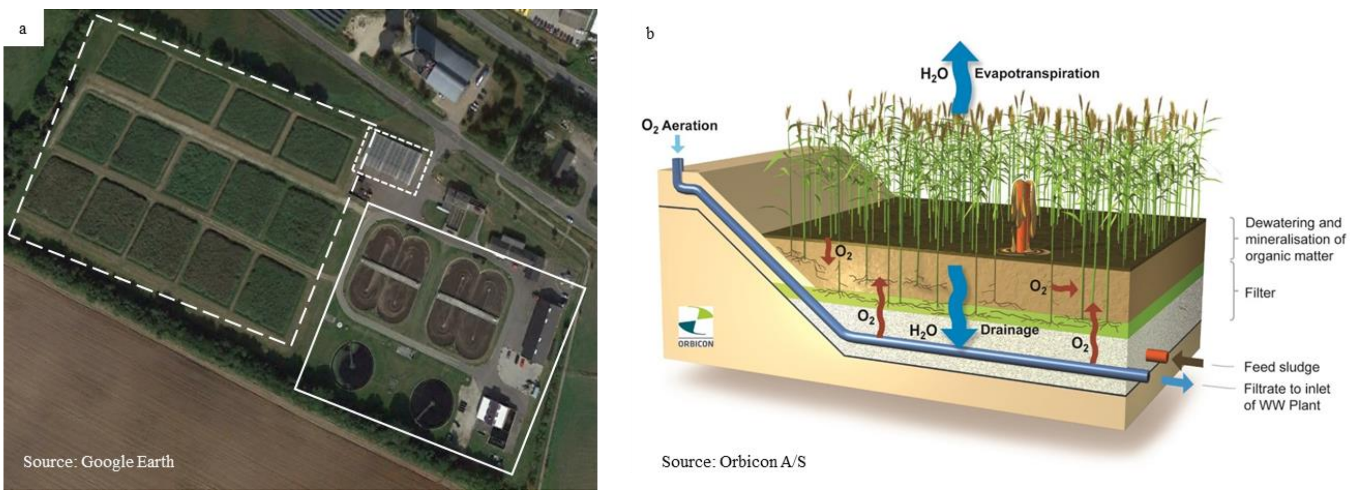

Figure 2. a) Aerial photo of an STRB system (coarse dashes) and an SPA (fine dashes) at Helsinge WWTP (full-line) (Denmark). The STRB system consists of 14 beds. The SPA is covered by a greenhouse roof. B) Schematic cross-section of a bed in an STRB system.

The loading procedure for STRB systems involves one bed at a time being loaded with sludge, while all the other beds in the system rest. During a “loading period”, a bed is loaded on a daily basis. Commonly, this takes two to seven days; however, the time can vary depending on the required treatment capacity, the number of beds, etc. When a bed has been subjected to a full loading period, the loading shifts to the next bed in the row and so on. The time span between two loading periods is called a “resting period”, the duration of which is equal to the number of days it takes before all other beds have been loaded. For example, if an STRB system contains 10 beds, and a loading period for each bed takes five days, the resting period for each bed will be 45 days. The basic idea is that all beds in an STRB system are subjected to loading and resting periods of equal duration. However, in reality, some of them occasionally need to be excluded from the loading scheme for a period, meaning that the other beds must receive more sludge to balance the need for treatment. Excluding specific beds from the loading scheme could be due to planned activities such as an upcoming emptying event, requiring that the beds chosen to be emptied rest longer than usual, or unplanned activities, such as the restoration of beds accidentally subjected to overloading.

The daily operation of an STRB system is relatively simple and requires only a modest input of staff working hours (Nielsen & Bruun 2015). Nonetheless, it is crucial to the treatment process that the loading and resting periods are of an appropriate duration, that the batches of sludge are of appropriate size and that the fed sludge is of appropriate composition. Sludge treatment reed bed systems are mainly used for treating surplus activated sludge (SAS), though the treatment procedure is also applicable to other sludge types, e.g. aquacultural sludge (Summerfelt *et al.* 1999) waterworks sludge (Nielsen & Cooper 2011) or septage (Troesch *et al.* 2009). Nevertheless, even though STRB systems are capable of treating sludge of different origins, some characteristics of the sludge should fulfil specific requirements, in order to obtain optimal dewatering efficiency. Fat and oil contents, for instance, should be lower than 5,000 and 2,000 $mg \cdot kg\ DS^{-1}$, respectively, and loss on ignition (LOI) should be a maximum 65% of DS (Nielsen 2011). If the feed sludge does not fulfil these requirements, the STRB system could be overloaded, a condition that, if not treated in time, leads to operational problems. If the sludge meets these requirements, the final sludge product can achieve a DS content of 20-37% (Nielsen 2011) under Danish climate conditions.

All beds in an STRB system are constructed as single units. Figure 2b shows a schematic cross-section of a single bed. The bottom and lateral surfaces are lined to prevent water, nutrients and pollutants from leaching out and into the surroundings (Nielsen 2003). A filter layer, in which two distinct pipe systems are embedded, covers the bottom, and then the loading pipe system leads sludge from the WWTP to the beds. The distributional parts of the pipes protrude vertically from the bottom of the beds, as shown in Figure 3a, and are designed to minimise gas stripping and to help distribute the sludge equally on the surface of the sludge residue.

Figure 3.. a) Active loading pipes in an STRB system. The pipes protrude vertically from the sludge residue and ensure the even distribution of sludge over the surface. b) The reject water system underneath the beds is open to the atmosphere (pipes protruding from the ground), thus allowing oxygen to enter the filter layer and the lower parts of the sludge residue.

The reject water pipe system collects water draining from the sludge residue; furthermore, it has an additional function as an aeration system, as it opens up to the atmosphere and allows atmospheric air to enter the filter layer and the lower parts of the accumulated sludge (Figure 3b). Within the first 24 hours after loading, approximately 95% of the water contained by the feed sludge drains off and is collected by the reject water system, which then returns it to the inlet of the WWTP, where it is treated along with incoming wastewater. Afterwards, the draining rate decreases, as most of the water held by the pore space of the sludge residue has now drained off.

2.2 Dewatering and mineralisation

Dewatering of the sludge residue continues during the resting period, now mainly driven by evaporation from the surface of the sludge residue and evapotranspiration from the reeds growing therein (Nielsen 1993). This enhancement of the dewatering process adds to the volume reduction of the sludge residue and prevents the sludge residue matrix from being water-locked.

The beds are planted with common reed (Figure 4a), which grows in the sludge residue and enhances the dewatering process via evapotranspiration and by creating cracks in the surface through which water can evaporate (Figure 4b) (Nielsen 2003).

Figure 4. a) The beds in an STRB system are planted with reeds, which are never harvested and follow their natural growth cycle. This photo was taken during summer, so the reeds stand tall and green. b) Their stems make openings in the sludge residue surface, allowing water to evaporate and oxygen to enter.

Even though common reed (*Phragmites australis*) is the plant species commonly used in an STRB system, other plant species have also been tested (Wang *et al.* 2010; Kengne *et al.* 2011; Uggetti *et al.* 2012a; Wu 2015).

During its yearlong residence in the beds, the sludge residue is not only dewatered, but also mineralised. The mineralisation of the organic parts of the sludge residue is driven by naturally occurring micro fauna, resembling those found in substrates of natural, reed-dominated habitats. I addition to roots, reeds grow long, hollow outgrowths, called rhizomes, which penetrate the substrate. Air leaks from the rhizomes into the pore space of the sludge residue matrix, creating aerobic microenvironments. The composition of the micro fauna present in the sludge residue is strongly dependent on the presence or absence of oxygen. The beds in an STRB system are designed to enhance a microenvironment dominated by aerobic microorganisms. Aerobic respiration of organic material produces CO_2, while anaerobic respiration produces CH_4 and CO_2.

Methane from biological processes related to wastewater and sludge treatment, however, is not considered climatically-neutral and has a GWP of 28 on a 100-year timescale (not including feedbacks) (IPCC 2014). Enhancing aerobic respiration of organic matter thereby limits the contribution to climate change. Furthermore, aerobic mineralisation is faster and more effective compared to anaerobic mineralisation (Vymazal *et al.* 1998). Carbon dioxide produced from biological processes related to wastewater and sludge treatment processes is classed as short-cycled carbon (fixed from and re-released into the atmosphere over 100 years) and thereby climate-neutral (IPCC 2007).

Even though the beds are designed to enhance aerobic microenvironments, anaerobic microenvironments will always be present. However, even though aerobic mineralisation is considered more effective in terms of degrading organic material, the microorganisms restricted to anaerobic and oxygen-limited environments also carry out processes beneficial to the treatment process. Denitrifying microorganisms, for instance, converts nitrate (NO_3^-) to free nitrogen (N_2), which reduces the amount of NO_3^- in the sludge residue and thereby its potential for eutrophication when it is eventually applied to land. However, denitrification can also produce N_2O as a by-product. Nitrous oxide is a potent greenhouse gas with a GWP of 265 (not including feedbacks) (IPCC 2014) and is therefore of major concern in relation to climate change.

The reeds also contribute to the conversion of N-based nutrients by taking up ammonium (NH_4^+) and NO_3^- as an N-supply through their roots and incorporating it into their biomass. However, N taken up by the reeds does not truly leave the system, since the reeds are never harvested from the beds but wither and die, due to their natural life cycle. The dead plant material is eventually incorporated into the sludge residue, where it is subjected to mineralisation processes along with the part of the sludge residue originating from the feed sludge; hence, any N taken up by the reeds is eventually returned to the sludge residue, albeit now built into organic matter.

2.3 Excavation and post-treatment

The beds in an STRB system can be loaded with sludge over several years, without being emptied. As more sludge is loaded into the beds, new layers cover the upper layers, resulting in the build-up of a body of dewatered and partly mineralised residue (Figure 5b). As a rule of thumb, 10 cm of sludge residue corresponds to one year's supply of sludge, the deepest layers being the oldest and the upper layers being the youngest. The older layers are more mineralised and dewatered, due to longer residence in the system (Pempkowiak & Obarska-Pempkowiak 2002; Kolecka & Obarska-Pempkowiak 2008; Peruzzi *et al.* 2013; Nielsen & Bruun 2015; Larsen *et al.* 2017a). Thus, time is an important aspect of STRB systems, whereas most treatment technologies based on mechanical dewatering and subsequent storage have relatively short time spans (approximately one year).

Figure 5. a**) Excavation of sludge residue and reeds from a bed in an STRB system. b) As sludge is loaded into the beds over several years, a body of residue builds up. The lowermost layers have resided in the bed the longest. As a rule of thumb, 10 cm of sludge residue corresponds to the build up of one year's supply of sludge.**

If an STRB system is operated according to design recommendations (Nielsen 2012), each bed can commonly receive sludge for 8 to 12 years before it must be emptied (Figure 5a). The loading scheme should be timed to ensure that not all beds require emptying in the same year, and so usually one or two beds are emptied at a time. When the reeds, which are excavated together with the sludge residue, have regrown, the emptied bed can run another 8-12 years before it must be emptied; this emptying cycle can be repeated three or four times before the bed must be renewed. Prior to an emptying event, the sludge residue in the target bed should rest for 4 to 12 months. The excavated sludge residue is commonly applied as fertiliser to agricultural land, normally during the autumn months. Emptying is done immediately before application and the excavated sludge residue transported directly to the application site. However, in recent years, a new procedure has been employed for some STRB systems, whereby emptying is undertaken during spring, before the initiation of the growth season. The excavated sludge residue is then transferred to an SPA, situated at the WWTP, where it is stored for 3 to 6 months until subsequent land application in autumn. This approach has the advantage that rhizomes in the sludge residue left in the bed will start regrowing shortly after the emptying process.

When emptying is undertaken during autumn, the reeds will not regrow until the subsequent spring. Emptying in spring thereby shortens the time required for the reeds to regrow by 6 to 8 months, which means that the STRB system returns faster to normal treatment capacity.

The original design of SPAs was very simple, i.e. an outdoor area on which sludge residue is piled (Figure 6a). Even though leaching of water and nutrients from sludge residue piled on an SPA is very limited, its base is sealed by a water impermeable membrane, to prevent water, nutrients, metals and xenobiotics from leaching into the environment. Later on, a shelter consisting of a greenhouse roof was added to the SPA design (Figure 6b). When sludge residue, including reeds, is excavated and piled on an SPA, the different layers of sludge residue are mixed into a homogeneous mass and aerated, due to excavation activity, which enhances evaporation and aerobic microbial activity. The reeds are crushed and mixed into the sludge residue, returning the N taken up during the treatment period in the STRB system to the sludge residue incorporated into organic matter, which now can be mineralised. Furthermore, the reeds provide a coarser texture to the sludge residue.

Figure 6: a) Sludge residue piled on the stockpile area at Himmark WWTP in 2014. b) Sludge residue piled at the stockpile area at Helsinge WWTP in 2016. The area is covered by a greenhouse roof, and greenhouse walls on two sides.

The final sludge product produced by STRB systems is known to meet the threshold values for use on agricultural land, as stated by the Danish Environmental Protection Agency (86/278/EEC EU Directive ENV) and the European Sewage Sludge Directive (BEK No. 86/278/EEC EU Directive (Nielsen & Bruun 2015; Larsen *et al.* 2017a), regardless of whether or not the treatment process has been supplemented with post-treatment on an SPA.

3 Methodology

In this chapter, the reference WWTPs and the technology involved in the fieldwork, laboratory work and data processing underlying the data generation and the LCA are presented. Fieldwork, laboratory work and data processing related to the three research areas (defined in chapter 1.2.2) are presented in three separate sections. In the last section, the technology and inventory data used for the LCA model are presented.

3.1 Reference wastewater treatment plants

Three reference STRB systems located at Helsinge WWTP, Himmark WWTP and Stenlille WWTP, were chosen as experimental sites for the practical data generation activities.

Reference STRB 1: Helsinge

The STRB system at Helsinge WWTP, Denmark (56°01’15N; 12°19,49E), which annually treats sludge corresponding to approximately 25,000 PE, was established in 1996 (Table 1). Originally, this STRB system consisted of 10 beds, but in 2013, four new beds were added, making 14 in total (Figure 2a).

Until 2010, Helsinge STRB system was loaded with a mixture of two sludge types, namely SAS produced by the plant’s biological-mechanical treatment line, and SAS produced at 14 smaller WWTPs in the area. Due to storage in tanks prior to transportation, the SAS produced at the smaller WWTPs is more concentrated and anaerobic compared to the SAS produced at Helsinge WWTP. Hence, this sludge mixture had a different composition compared to the SAS produced at Helsinge WWTP. Since 2010, Helsinge STRB system has been loaded mainly with pure SAS from Helsinge WWTP. However, the change of sludge type in 2010 means that the deepest layers (40-50 cm) of sludge residue in the beds originate from the mixed sludge type, while the uppermost layers originate from SAS produced at Helsinge WWTP, occasionally mixed with concentrated anaerobic SAS.

Today, SAS produced at the 14 smaller WWTPs is dewatered on the decanter centrifuge at Helsinge WWTP. The centrifuge is run daily, producing dewatered sludge that is stored in an open container, which is emptied once a week, and then transported to an external sludge storage facility in Store Merløse, Denmark.

Table 1: System characteristics and characterisation of feed sludge for STRB systems associated with the WWTPs in Helsinge, Himmark and Stenlille, Denmark. Data source: Orbicon A/S.

System characteristics	Helsinge	Himmark	Stenlille
Year of construction	1996	2003	2006
Population equivalent served (PE.)	25,000	18,000	10,500
Capacity (tons ds y^{-1})	630	350	175
Number of beds	14	10	8
Single bed area (m^2)	1,050	700	365
Total bed area (m^2)	14,700	7,000	2,920
Loading rate (kg ds m^{-2} y^{-1}) – Dim.	60	50	60
Loading rate (kg ds $m^{-2}y^{-1}$) - Real	45	36 - 46	23 - 29
Loading/resting (days)	4-7/40-60	4-7/40-70	3-4/56-56
Feed sludge	**Helsinge**	**Himmark**	**Stenlille**
Sludge type	SAS/Digested sludge	SAS mix.	SAS
Dry solid (% of WW)	0.6 - 0.8	0.6-0.8	0.4
Loss on ignition (% DS)	45 - 65	40 – 55	55-65
Sludge age (aerobic days)	20 - 25	20 - 25	20 - 25
Phosphorus removal	PIX/Fe	PAX/PIX	PIX/Fe

To obtain data for the LCA on the dewatering efficiency of the centrifuge, it was arranged in February 2015 that one day's production of the SAS produced at Helsinge WWTP, which is commonly loaded into an STRB system, would be redirected to the centrifuge and dewatered.

In 2012 and 2013, an SPA was constructed at Helsinge WWTP (Figure 6b) with a total area of 1675 m^2. A greenhouse roof covers 800 m^2, which is the area required for the post-treatment of the amount of sludge residue contained by one bed. Later, in 2016, greenhouse walls covering two sides were added. The roof and walls enhance solar drying of the sludge residue, which leads to a considerable loss of water and organic material to evaporation and mineralisation processes, respectively.

Reference STRB 2: Himmark

The STRB system at Himmark WWTP, Denmark (55°2'44"N 9°45'55"E), was established in 2003 and treats sludge annually, corresponding to 18,000 PE (Table 1). The feed sludge is a mixture of SAS, produced by the WWTP's mechanical-biological wastewater treatment line, and digested sludge from a mesophilic digester. In 2010, an SPA was constructed (Figure 6a) but without a greenhouse roof or walls.

Reference STRB 3: Stenlille

The STRB system at Stenlille WWTP, Denmark (55°3'25''N 11°34'34''E), was established in 2006 and annually treats sludge corresponding to 10,500 PE (Table 1). The feed sludge is SAS, produced by the WWTPs mechanical-biological WW treatment line. The WWTP has no SPA.

3.2 Research area 1: Quantification of biological gas emissions from sludge treatment and storage

The focus of Research area 1 centred on investigating gas emissions caused by biological activity in sludge subjected to treatment in STRB systems, post-treatment at SPAs and storage subsequent to mechanical dewatering.

Seasonal gas emission rates from STRB system

Fieldwork

To obtain data on CO_2, CH_4 and N_2O emissions covering all four seasons of the year, four measuring periods, each covering one season, were scheduled. As gas emission activity in sludge residue is affected by loading state (Olsson *et al.* 2014; Larsen *et al.* 2017b), each measuring period was scheduled to cover an entire resting period. Table 2 provides an overview of the measuring periods.

Table 2. Overview of the measuring periods during which gas emissions were recorded at Helsinge STRB system.

Season	Loading (number of days)	Resting (number of days)	Measuring periods
Spring	4	43	March 4 - April 14, 2015
Summer	4	23	July 14 - Aug 5, 2015
Autumn	4	37	Oct 14 - Nov19, 2014
Winter	4	37	Jan25 - Feb 29, 2016

Bed 4 in the STRB system at Helsinge WWTP was chosen to host all measuring activities. In this bed, locations for seven measuring stations were chosen. The surface area of the bed was mapped in a 32 x 32 m grid and the position of the seven measuring stations defined (Figure 7).

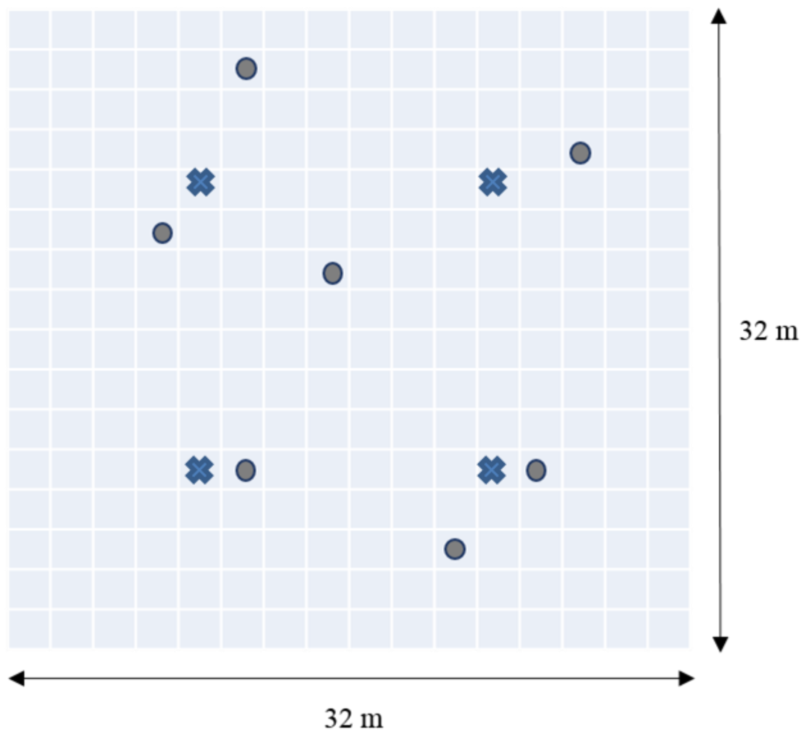

Figure 7. Schematic overview of Bed 4 in the Helsinge STRB system. The circles specify the location of the seven surface flux chambers, and the crosses note the location of loading pipes.

At these stations, static surface flux chambers (one at each station) were installed prior to each measuring period. The paths leading to the measuring stations were enforced by wooden boardwalks. By the end of each measuring period, flux chambers and boardwalks were removed from the bed, in order to disturb reeds and sludge residue as little as possible.

As the measuring periods were timed to cover resting periods, the bed was always subjected to a loading period prior to the start of the measuring period. On the first day of a measuring period (and thereby the first day in a resting period), static surface flux chambers were installed at the seven measuring stations. The chambers were constructed according to the guidelines provided by Livingston and Hutchinson (1995) (Figure 8a); this design had been applied previously, in order to measure gas emissions from STRB systems, with successful outcomes (Uggetti *et al.* (2012b); Olsson *et al.* (2014)). The chambers used in the present study were constructed from plastic barrels (Figure 8b), the bottoms of which were cut out, leaving them approximately 40 - 45 cm tall. The barrels came with tightly fitting lids, which were equipped with septas, thereby making it possible to insert a thermometer and a rubber tube for gas extraction. A chamber was installed at a measuring station by pushing the bottom edge approximately 10 cm into the sludge residue, to prevent gas from leaking from the sides. Prior to installation, the reeds were cut to approximately 20 cm in height, making it possible to fit the chamber around them (Figure 8a). Grünfeld and Brix (1999) found that cutting reeds did not affect gas emission rates significantly; hence, it was assumed that the cutting would have no effect on the results. On the inner side of the lids a fan was mounted to ensure the mixing of the gases emitted into the chamber from the sludge residue.

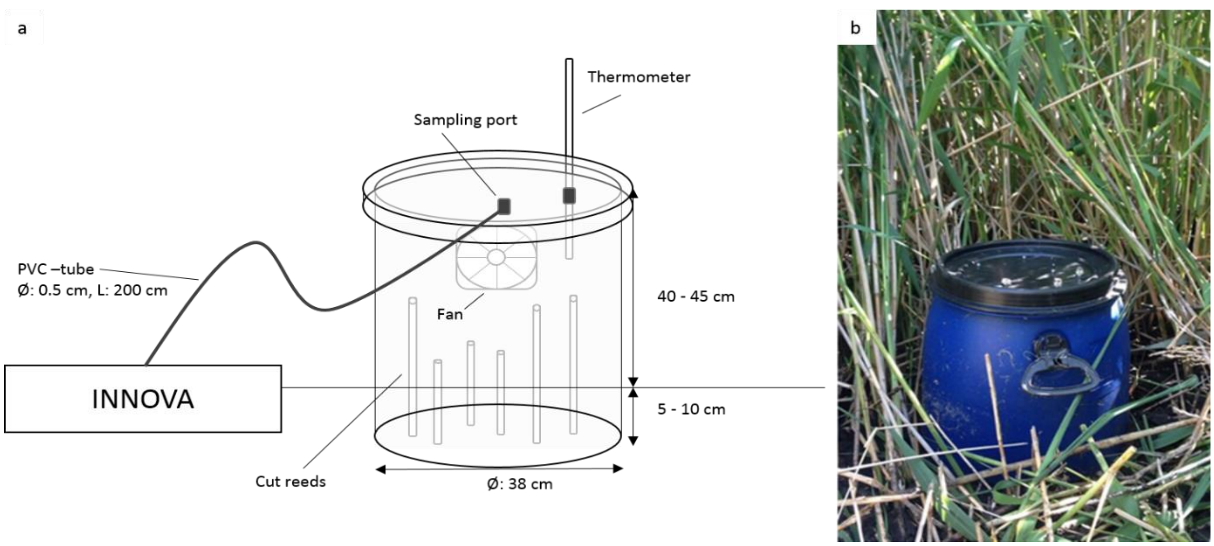

Figure 8. Design of a static surface flux chamber, based on guidelines provided by Livingston and Hutchinson (1995). b) Static surface flux chamber mounted in an STRB system at Helsinge STRB system.

All measuring periods included 10 measuring dates on each of which fluxes of CO_2, CH_4 and N_2O were recorded from each of the seven chambers. The lids were only mounted on the chambers during measuring activity; otherwise, they were left open. For all measuring periods, the first measuring date was scheduled for the second day of the resting period.

On a measuring date, gas emissions from each of the seven measuring stations were recorded once, one at a time. Measuring activity was initiated at 9-10 am and finished around two hours later. A chamber was prepared for measuring activity by mounting the lid with the fan turned on two minutes before initiation of recording. Gas accumulation in the headspace of the chamber was recorded by using a mobile photoacoustic gas monitor (Gas Monitor INNOVA 1312 (Innova AirTech Instruments) connected to the chamber by a 200 cm-long PVC tube (Ø 0.5 cm). The INNOVA was set up to extract a gas sample once a minute, over five to ten minutes. The temperature in the headspace of the chamber was noted and the equipment moved to the next chamber until all chambers had been visited.

Data processing

For all measuring dates, gas emission rates ($g \cdot m^{-2} \cdot d^{-1}$) for the individual flux chambers were calculated as the linear increase of the gas concentration measured in the chamber. Regression lines with an r^2 value < 0.8 were excluded from the datasets. For each measuring date and each gas species, *a daily average gas emission rate* representative of the whole bed was calculated by applying the Kriging approach (Cressie 1990) to the emission rates measured from the seven measuring stations. These calculations were undertaken using SURFER® 13 (Golden Software, Inc., Colorado, USA) software.

Next, *an average seasonal emission rate* was calculated for each of the gas species. These calculations were based on average daily emission rates, calculated as described above. The average seasonal emission rates were calculated as temporal weighted averages (Equation 1), with *x* representing daily average gas emission rates obtained during the specific seasons, *w* representing the number of days passed since the last measurement and *n* representing the number of measurement dates.

$$\bar{x} = \frac{\sum_{i=1}^{n}(x_i \cdot w_i)}{\sum_{i=1}^{n} x_i} \qquad \text{(Equation.1)}$$

To calculate comparable average seasonal emission rates, all measuring periods were scaled up or down to cover 42 days, assuming that the emission rates recorded on the last day of a measuring period were representative of the period between the last measuring day and day 42. All resting periods were initiated by a four-day loading period. Emissions during the loading periods were accounted for by assuming that gas emission rates on a loading day were the same as on the second day of the subsequent resting period. Finally, we calculated *an average, annual emission rate* for each gas species, assuming that one season corresponded to 91.25 days.

Seasonal variations in the emission rates were tested by a one-way ANOVA test and converted to CO_2 equivalents, in order to calculate the annual contribution to climate change (Larsen *et al.* 2017c).

Gas emission rates from post-treatment at SPA

Gas emission rates representing the post-treatment of sludge residue at an SPA were recorded from May 2015 until August 2015. Five measuring stations were chosen and five static surface flux chambers installed as described for an STRB system (section 3.2. "Seasonal gas emission rates from an STRB system"). The first measuring date was scheduled the day after installing the chambers and followed by four measuring dates distributed over the post-treatment period, the last date being the day before the sludge residue was excavated from the SPA. The sludge residue was transported to the land application site immediately after excavation. On the measuring dates, the procedure described for an STRB system was followed. Average daily emission rates ($g \cdot m^{-2} \cdot d^{-1}$) of CO_2, CH_4 and N_2O were calculated, also as described for an STRB system.

Helsinge WWTP's SPA was first used in 2015. The greenhouse roof was present, but its greenhouse walls had not been constructed. As post-treatment at the SPA at that time was a new procedure, a practical procedure was not fully optimal. The SPA had been constructed to support the post-treatment of an amount of sludge residue contained by one bed; however, two beds were emptied, and the sludge residue was heavily packed on the SPA in an attempt to fit everything under the one roof, with some matter left outside, due to space issues. Even though sludge residue samples collected at the beginning and at the end of the post-treatment period revealed that they had been dewatered and mineralised further during the post-treatment period, operational errors presumably prevented the treatment process from reaching its full potential. Indeed, in 2016, sludge residue from one bed was excavated after a 12-month resting period and piled (not packed) at the SPA. The practical procedure was optimised and the greenhouse walls added to the design, resulting in a considerably higher loss of water and organic matter during the post-treatment period. However, we did not manage to record gas emissions in 2016. Therefore, data recorded during 2015 were used in the LCA.

Gas emission rates from storing dewatered sludge

Sludge dewatered at the centrifuge housed at Helsinge WWTP is stored in a container (L: 5 m, W: 3 m, H: 1.5 m) immediately after dewatering. The sludge is dewatered on daily basis, and the container can hold one week's production before it must be emptied and the dewatered sludge transferred to an external storage facility in Store Merløse, Denmark. To record gas emissions, in October 2015, a container was filled with dewatered sludge (originating from the 14 smaller WWTPs mentioned in chapter 3.1, "Helsinge WWTP") over one week and moved to an outside, roof-covered, area. After moving the container, five static surface flux chambers were mounted in the dewatered sludge, distributed as evenly as possible over the surface. The following day, emissions of CO_2, CH_4 and N_2O were recorded with the same procedure as described for an STRB system (chapter 3.2, "Seasonal gas emission rates from STRB system"). The recording procedure was repeated four days later, and based on the data recorded at these two measuring dates, average, daily gas emissions rates ($g \cdot m^{-2} \cdot d^{-1}$) representing one week of on-site storage of mechanically dewatered sludge at Helsinge WWTP were calculated.

Afterwards, the container holding the dewatered sludge was left untouched, and gas emissions recorded again after 33 and 100 days. Daily emission rates of CO_2, CH_4 and N_2O, representing the storage of dewatered sludge at an external sludge storage facility, were based on data obtained from the container, assuming that the average daily emission rates based on the four measuring dates were representative of a whole year. As the mechanically dewatered sludge stored at the external storage facility in Store Merløse is never turned or moved during storage time, and no other activities are done to enhance the dewatering or mineralisation of organic material, this was assumed an acceptable approach.

Originally, the plan was that the gas emission measurements would be carried out on mechanically dewatered sludge originating from Helsinge WWTP's internal wastewater treatment line, not from the 14 smaller WWTPs. However, redirecting the sludge produced at Helsinge WWTP from being loaded into the STRB system for mechanical dewatering was very time consuming for staff working at Helsinge WWTP, meaning that the plan had to be reconsidered. As the sludge transferred from the smaller WWTPs also originated from mechanical-biological wastewater treatment lines, and that a share of this sludge sometimes is mixed into the SAS produced at Helsinge WWTP, before being loaded into an STRB system, it was assumed that gas emission rates representing the storage of sludge dewatered on a centrifuge originated from sludge transferred from the 14 smaller WWTPs.

Change in gas composition in sludge residue between loadings

Between September and October 2015, the percentage gas composition in the pore space and surface fluxes of sludge residue residing in Bed 5 at Helsinge was recorded. The measuring dates were timed to follow a resting period, as the intention was to follow how the percentage gas composition in the pore space at different depths changes as the sludge residue is gradually dewatered after a loading period. The results are not to be used in the LCA but added to knowledge on temporal gas emission dynamics related to the loading scheme (Larsen *et al.* 2017b).

3.3 Material and substance flow analysis

To establish substance flows for the treatment of sludge in STRB systems, post-treatment at SPAs and mechanical dewatering and storage, samples of sludge, reject water, sludge residue and mechanically dewatered sludge were collected and their composition analysed.

STRB system substance flow

Fieldwork

The samples of sludge, reject water and sludge residue, underlying the substance flow analysis for STRB systems, were all taken from Bed no 9 at Himmark's STRB system in March 2014. The samples were taken on the last day of a resting period (lasting 42 days), while feed sludge and reject water were sampled the following day.

As the sludge residue found at different depths in a bed has resided in a bed for different numbers of years (Figure 5b), any changes in concentrations of substances related to treatment time can be revealed by analysing separately the composition of the distinct layers.

In order to sample sludge residue, eight locations were randomly chosen in the bed. At each location, vertical cores of sludge residue were collected with an iron core sampler (Ø 2, 5cm). On the sampling day, the height of the sludge residue in the chosen bed was 100 cm. However, as approximately 10 cm of sludge residue is left in the beds when these are emptied, the difference in age between the deepest layer in a bed and the overlying layer could be more than one year. Therefore, the cores were taken to a depth of 90 cm. The cores were cut into sections of 10 cm (Figure 9), and sludge residue originating from the distinct sections was pooled together in large plastic bags: all sections representing samples taken from 0-10 cm were pooled together, all sections representing samples from 10-20 cm were pooled together and so on. Inside the bags, the samples were kneaded by hand into a homogeneous mass.

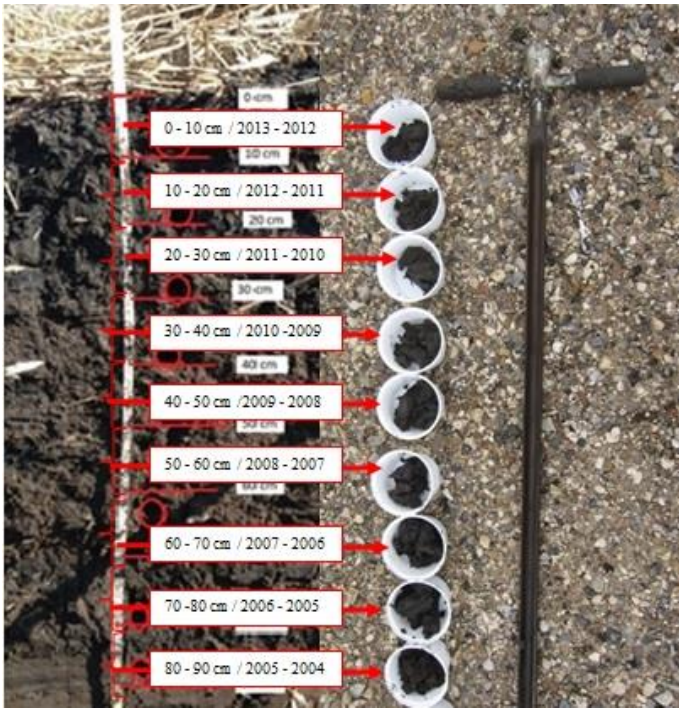

Figure 9. In order to collected sludge residue for characterisation, vertical cores of sludge residue were retrieved by a core sampler (right). The labels indicate when the different layers were created. The layers were analysed seperately.

The following day, feed sludge and reject water samples were collected. As reject water continues to drain from the sludge residue 24 hours after loading, a composite sample was created by taking 1 L of reject water every time 8 m^3 of reject water had had passed through the reject water system, finally mixing these together.

Laboratory analysis

All samples (sludge, reject water and sludge residue) were analysed for contents of dry solids (DS), volatile solids (VS), total carbon (TC), total nitrogen (TN), NO_3^-, NH_4^+, phosphorous (P), potassium (K), calcium (Ca), iron (Fe), manganese (Mn), chromium (Cr), nickel (Ni), cobber (Cu), zinc (Zn), cadmium (Cd) and lead (Pb). Contents of DS and VS were analysed by drying and igniting the samples. Contents of NO_3^+ and NH_4^+ were analysed by employing an automated ion analyser (FIAstar 5000 flow injection analyser (Foss Analytical, Denmark)). Contents of P and mineral elements (Cr, Ca, Mn, Fe, Ni, Cu, Zn, Cd, Pb, and K) were analysed by an ICP-OES (Optima 2000 DV, Perkin Elmer, USA). Total N and TC were analysed with a CHN analyser (NA2000 Fisons Instruments, Italy).

Data processing

The uppermost layer of sludge residue, sampled from 0 to 10 cm in depth, has resided in the bed for 0 to 1 years, while the lowermost layer, sampled from 80 to 90 cm depth, has resided in the bed for 8 to 9 years (Figure 9). Based on the concentrations of VS, TC and TN in residue of different ages, the percentage amounts of these substances mineralised each year were calculated. As fresh sludge contains a large amount of readily degradable organic matter, the percentage amounts mineralised during the first year were much greater compared to the amount mineralised in subsequent years. Indeed, for VS, TC and TN, the percentage amounts mineralised during the first year were 57%, 54% and 52%, respectively, while the average annual percentage amounts mineralised for the subsequent eight years were 3%, 3% and 5%, respectively. Annual losses of DS were calculated by assuming that the amount of DS lost corresponded to the amount of VS mineralised during the corresponding year. Based on the percentage concentrations of water and DS obtained by analysing sludge residue of different ages, the percentage amounts of water lost each year were now calculated.

Based on these annual percentage mineralisation rates, the accumulation of VS, TC, TN, DS and water in the body of sludge residue built up in the bed over nine years was calculated.

These calculations were extrapolated to cover 12 years of accumulation. Based on compositional data on sludge loaded into the system between 2004 and 2013, the total amount of each substance supplied by the sludge during 12 years was estimated. From these results, the percentage share of each substance mineralised, accumulated or lost to reject water, related to the total input from the sludge during the entire treatment period (12 years), was now calculated. For water, the percentage amount lost to evapotranspiration was calculated.

For P, K and metals, it was assumed that these substances were only lost to reject water. Indeed, they are taken up by reeds, to a larger or smaller extent; however, as the reeds are never removed from the system but are eventually incorporated into the sludge residue, substances accumulated in these reeds eventually return to the sludge residue, too.

Substance flow of the SPA

Samples of sludge residue were collected from the SPA at Helsinge WWTP in May 2015 (one week after it was excavated from the beds) and in August 2015 (immediately before excavation and transfer to a land application site) on the same days as the gas emissions measurements, described in Chapter 3.2, “Gas emission rates from post-treatment at SPA”, were undertaken. Samples were collected with a core sampler, though the cores were not divided into depth sections in the same way as the samples collected from the STRB system but homogenised into a composite sample. The concentrations of different substances in the composite samples collected at the beginning and end of the post-treatment period were analysed according to the same laboratory procedures as described for the STRB system in the previous chapter. Based on the concentrations identified, the percentage shares of water, VS, TC, TN and DS lost during post-treatment were calculated. As no water leached from the SPA, it was assumed that no substances had been lost to leaching, meaning that the amounts of P, K and metals held by the sludge residue remained constant during the post-treatment period.

Substance flow following the storage of mechanically dewatered sludge

In February 2015, a day’s production of SAS produced at Helsinge WWTPs internal WW treatment line was redirected to the centrifuge and dewatered. During this event, sludge, reject water and dewatered sludge were collected from the centrifuging process. During the entire centrifuging process (3 hours), a sample of reject water and one of dewatered sludge were collected every 15 min, with 12 samples in total of each substance. For both reject water and dewatered sludge, the 12 samples were mixed into one composite sample. All samples were analysed according to the same lab procedures as described for the samples collected at the Himmark STRB system.

The gas emission rates of CO_2, CH_4 and N_2O calculated in Chapter 3.2, “Gas emission rates from storage of dewatered sludge”, were used to estimate the percentage loss of C, N and VS during on-site storage in the container and external storage at the external sludge storage facility. The loss of water to evaporation during on-site storage was assumed negligible. The loss of water to evaporation during external storage was estimated by using data on the water content found in dewatered sludge before and after 200 days of storage, as published in a report by the Ministry of Environment and Food of Denmark (Miljø- og Fødevareministeriet (2000)). As dewatered sludge stored at the external storage facility in Store Merløse is not turned, moved etc. during the storage period, nor any other activities done to enhance mineralisation or evaporation, the data and calculations based on storage in a container are assumed representative for storage at the external storage facility. Finally, as no water leached from the sludge residue during storage, it was further assumed that no P, K and metals had left the system.

3.4 Emissions related to the land application of sludge

To determine emissions related to the application of sludge to land, samples of sludge residue from STRB systems and mechanically dewatered sludge were collected, mixed with soil and incubated in the laboratory for about 160 days. During incubation, emissions of CO_2 and N_2O, and the accumulation of NH_4^+ and NO_3^-, were measured and the data used for simulating emissions over a longer time frame of 100 years.

Collection of samples for incubation

Sludge residue representing treatment in STRB systems

Samples of sludge residue for the incubation experiment were collected from STRB systems at Helsinge, Himmark and Stenlille WWTP. The samples were collected with a core sampler and divided into fractions of 10 cm each, according to the same procedure described in Chapter 3.3, "Substance flow of STRB system". Samples originating from Himmark STRB system were collected along with those to be used in the substance flow analysis and thereby included nine samples representing one to nine years of treatment (Chapter 3.3, "Substance flow of STRB system"). In addition to these samples, a composite sample was made by homogenising all layers of sludge residue retrieved via core sampling. When sludge residue is excavated from a bed, it is extensively mixed due to the excavation activity; thereby, the composite sample represents the composition of the sludge residue as applied to land. From STRB systems at Helsinge and Stenlille WWTP, samples of sludge residue were collected in March and April 2014. The bed chosen for sampling at Helsinge STRB system (no 4, the same as used for recording the gas emissions) contained 100 cm of sludge residue, representing one to ten years of treatment. Stenlille, on the other hand, only contained 40 cm of sludge residue, representing one to four years of treatment.

Sludge residue representing post-treatment in an SPA

Samples of sludge residue representing post-treatment at an SPA were collected from the SPA at Himmark WWTP in August 2014 after five months of treatment. In 2014, the SPA at Helsinge WWTP was not yet in operation; the first period of post-treatment ran in March to August 2015. As the incubation set-up simulating application to land must run for 160 days (five months), we did not manage to include the samples collected from Helsinge SPA in 2015 or 2016 in our incubation set-up, meaning that emissions related to land application from these samples could not be obtained. The SPA at Himmark WWTP is not covered by a greenhouse roof or walls (Figure 6a); therefore, the evaporation of water and mineralisation of organic matter during the post-treatment is less efficient compared to post-treatment at Helsinge's SPA. Hence, emissions related to land application from sludge residue subjected to post-treatment at an SPA may not reflect the best performance of the technology, albeit the results obtained are well representative for the original SPA design. Furthermore, no other data on emissions from sludge residue subjected to post-treatment at an SPA related to land application currently exist, so emissions in relation to land applications derived from these samples are the most representative for use in the LCA.

Sludge residue representing dewatering via a centrifuge

A sample of dewatered sludge was derived from the composite sample of dewatered sludge collected at Helsinge WWTP in February 2015 (Chapter 3.3, "Substance flow of storage of mechanically dewatered sludge"). Hence, emissions related to land application from sludge dewatered on a centrifuge represent dewatered sludge that has not been stored. We did not manage to include a sample of dewatered sludge that had been stored in the incubation set-up. It is possible that emissions related to land application for the obtained sample would be larger compared to if the sample had been stored for one year, and the environmental impacts thereby overestimated. On the other hand, as nothing is done to enhance the evaporation of water or the mineralisation of organic matter from dewatered sludge during storage, it is also likely that emissions would not be affected by this storage, or they may be even higher.

Incubation experiments

To simulate application to land, samples of sludge residue and dewatered sludge were mixed with soil of a type commonly found at Danish land application sites (sandy loam), following which the mixture was incubated for 160 days. During the entire incubation period, the temperature was fixed at 15ºC, namely the average air temperature in Denmark during spring and summer. One week before initiating the incubation experiment, the soil was moistened and pre-incubated to activate microorganisms present in the soil. During incubation, emission rates of CO_2 and N_2O, and the accumulation of NO_3^- and NH_4^+, for all samples were recorded on specific dates.

To obtain data usable for an LCA, results of the incubation experiments must be further processed, which will be described in the following chapter. Data on CO_2 and N_2O emissions, and the accumulation of NH_4^+ and NO_3^-, obtained by incubating the samples of sludge residue collected at Himmark STRB system and SPA were chosen for further processing, thereby representing emissions related to land application from sludge residue subjected to treatment in an STRB system.

Data processing

When modelling environmental impacts from land application for use in an LCA, the emissions rates of included substances should cover a time period of 100 years. To achieve this aim, data obtained by incubating samples representing treatment in an STRB system (composite sample from Himmark STRB system), post-treatment at an SPA (composite sample from Himmark SPA) and dewatering on a centrifuge (composite sample from the centrifuge at Helsinge WWTP) were processed by using the agroecosystem software model DAISY (Abrahamsen & Hansen 2000; Hansen *et al.* 2012). This model takes a number of relevant conditions into account, such as crop rotation, agricultural procedures and climate. By applying the emission rates of CO_2 and N_2O, and the accumulation rates of NO_3^- and NH_4^+, obtained from the incubation experiment, gaseous emissions of N_2O, ammonia (NH_3, leaching of NO_3^- to groundwater and surface water, crop uptake of N and carbon sequestration by the soil years were calculated and extrapolated to cover 100 years.

3.5 Life cycle assessment of sludge treatment technologies

The LCA comparing STRB systems to mechanical treatment on a centrifuge and subsequent storage is presented in Larsen *et al.* (2017d). The manuscript and “Supplementary Information” (SI) of this paper include an extensive section addressing assumptions and the life cycle inventory and life cycle impact assessment (LCIA) underlying the LCA modelling, which will be used as a reference when describing the LCA technology in the coming section.

Principles of life cycle assessment

The idea behind the LCA approach is to evaluate the entire life cycle of one or more products or services. Each step in the production of the product or the service activity is defined and the environmental impact and/or economical expenses related to each step evaluated. The approach is used widely to compare the performance of different technologies serving the same purposes (e.g. treatment of sludge in STRB systems vs. mechanical treatment) and is therefore a useful decision-making tool.

LCA technology is defined in ISO 14040 and 14044 (ISO 2006). Overall, it involves four phases. The first phase defines the goal and scope of the LCA. Borders limiting the extent of included sub-processes should be carefully defined, especially if more scenarios are compared. Furthermore, a functional unit (FU) must be defined – a quantification of the service being delivered (e.g. what is the environmental impact of treating 1000 kg wet weight (WW) of sludge?) and must be the same for all scenarios compared within the LCA.

The second phase establishes a life cycle inventory, which defines inputs (e.g. fuel consumption) and outputs (e.g. CO_2 emissions related to fuel consumption) for all sub-processes.

The third phase is the LCIA, during which total impacts adding to various impact categories from the different scenarios are calculated by relating the LCI data to the FU.

In the final fourth phase, the results are interpreted and evaluated.

Goal, scope definition and functional unit

The goal of the LCA, as defined in section 1.2, was to compare the environmental performance of STRB systems with a conventional treatment technology based on dewatering on a centrifuge and then subsequent storage. The FU was defined as the treatment and disposal of 1000 kg wet weight (WW) of SAS with characteristics corresponding to SAS generated at Helsinge WWTP. The characteristics of this SAS are illustrated in Table 3. Information on the previous wastewater treatment process is to be found in Larsen *et al.* (2017d), section SI-1.

The LCA was modelled as an attributional LCA, meaning that the goal was to evaluate the environmental performance of the included scenarios in relation to a specific reference year. As a comparison, consequential LCAs seek to identify the future consequences of a defined change in the included scenarios; however, this is not relevant in the present LCA, as the aim was to evaluate the scenarios based on their present-day best performance, the reference year being 2016.

Table 3. Characterisation of the SAS produced by the biological wastewater treatment line at Helsinge WWTP.

Parameter		Parameter	
TS (% of WW)	0.6790	Cr (% of DW)	0.0023
LOI (% of WW)	61.483	Mn (% of DW)	0.0747
TN (% of DW)	3.9700	Fe (% of DW)	6.3970
TC (% of DW)	27.890	Ni (% of DW)	0.0022
NO_3^--N (% of DW)	0.000015153	Cu (% of DW)	0.0314
NH_4^+-N (% of DW)	0.000000001	Zn (% of DW)	0.0573
Mg (% of DW)	0.4234	Cd (% of DW)	0.0001
P (% of DW)	2.2900	Pb (% of DW)	0.0030
Ca (% of DW)	2.8255	K (% of DW)	0.3911

*DW = dry weight

The temporal horizon of the LCA is 100 years, meaning that the fate and environmental impact of the different output substances (greenhouse gases, etc.) during the subsequent 100 years after entering the environment was evaluated.

Three scenarios, all initiated by sludge entering the specific sludge treatment scenarios and terminated when the final sludge product is applied to the land, were defined. The sub-processes of each scenario are shown in Figure 10.

Scenario 1: Centrifuge and storage (S-CEN)

Sludge is dewatered on a conventional decanter centrifuge and immediately transferred to a container in which it is stored for one week (“on-site storage”). Afterwards, the dewatered sludge is transported 70 km by truck to an external sludge storage facility (“external storage”), where it is laid out on the floor in an enclosed storage building. The dewatered sludge is not moved or treated during storage. The storage facility continually receives dewatered sludge throughout the year, meaning that at the time of land application, some of it has been in storage for one year, while some has only been stored for a few days.

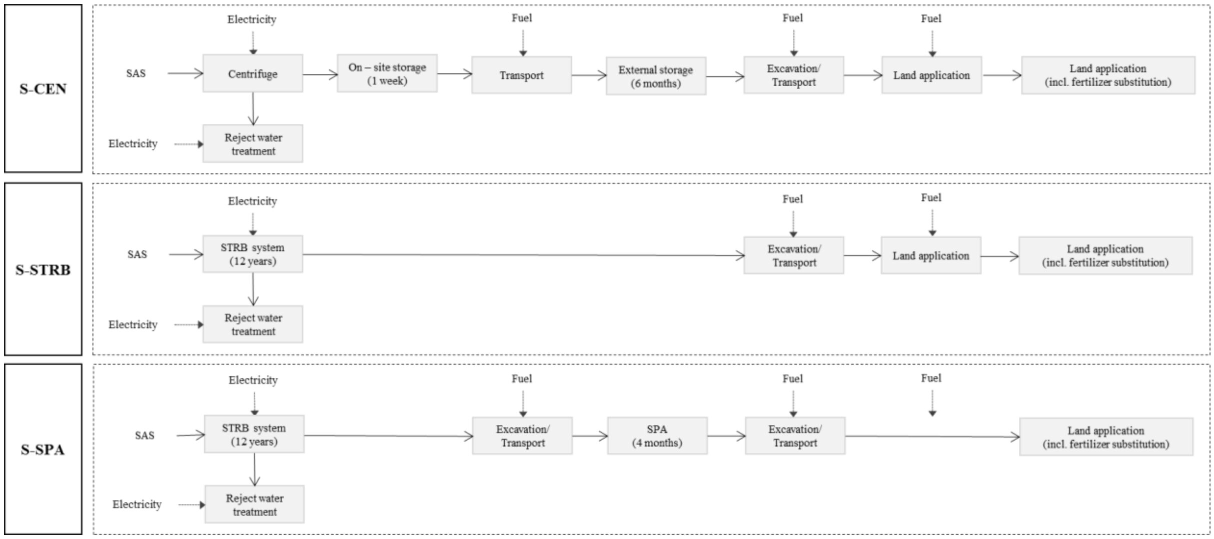

Figure 10. Unit processes for three sludge treatment scenarios: S-CEN (dewatering on centrifuge, one week of on-site storage in container and six months of external storage until land application), S-STRB (12 years of treatment in a STRB system, excavation in autumn and immediately application on land) and S-SPA (12 years of treatment in STRB and excavation in spring, followed by four months of post-treatment at a SPA).

Hence, the average storage time was defined as six months. At the time of land application, the stored sludge is excavated from the storage building by a skid steer excavator, transported 200 km by truck to a land application site and applied by a tractor.

Scenario 2: Sludge treatment reed bed system (S-STRB)

A bed in an STRB system is loaded with sludge over 12 years, resulting in the build-up of a body of sludge residue. After 12 years, the entire body of sludge residue (including reeds) is excavated by an excavator, transported 10 km by truck to a land application site and applied by a tractor.

Scenario 3: Sludge treatment reed bed system and stockpile area (S-SPA):

A bed in an STRB system is loaded with sludge over 12 years, resulting in the build-up of a body of sludge residue. After 12 years, the entire body of sludge residue (including reeds) is excavated by an excavator and transported 0.15 km by truck to the SPA. Here, it is laid out and undergoes four months of post-treatment. Finally, it is excavated from the SPA by a skid steer excavator, transported 10 km by truck to a land application site and then applied by a tractor.

The EASETECH model

LCA modelling was undertaken by using the EASETECH software model (Clavreul et al. 2014) (formerly known as EASEWASTE) developed by DTU Environment. This software models the flow of substances and related emissions through waste treatment scenarios defined by the user. Substance flows are based on inputs of characterised material fractions (e.g. sludge) and emission data provided by inventory databases, such as Ecoinvent or the database imbedded in the EASETECH software, or as defined by the user. EASETECH provides a row of template processes, making it possible for the user to model environmental emissions (e.g. gaseous emissions or leaching into terrestrial or aquatic environments) from waste treatment technologies (e.g. an STRB system) and activities related to the treatment process (e.g. transportation of treated sludge). Emissions are then processed according to the standards of the LCIA and the impacts sorted into different impact categories.

For the present LCA, an input material fraction was created based on the SAS from Helsinge WWTP, described in Table 3, and processes reflecting activities and related emissions in the included treatment scenarios (Figure 10), programmed by entering data on biological gas emissions, flow of substances and emissions related to land application, all of which were obtained from the data generation activities described in Chapters 3.2, 3.3 and 3.4.

Life Cycle Inventory

All operational data on energy consumption transport etc. for the different scenarios were provided by the Helsinge WWTP (Grib Vand A/S). All scenarios included the consumption of electricity, due to daily operations (pumping of sludge and reject water, running the centrifuge) and the consumption of fuel for excavation and transport. Prior to mechanical dewatering, polymer coagulant is added to the sludge; therefore, emissions related to the production of polymer coagulant were included in S-CEN. Data on emissions related to the use of heavy vehicles, electricity consumption and the production of polymer coagulant were taken from the international LCI database Ecoinvent (v. 3.3), which is included in the EASETECH software (v. 2.3.6) (described in the next chapter). The data used are presented in Larsen *et al*. (2017d), section SI-3.

In S-CEN, it was assumed that the final sludge product is transported 200 km to the land application site, while in S-STRB and S-SPA this distance was only 10 km. Sludge treated in a well-operated STRB system meets the threshold values for heavy metals and xenobiotics for land application of biosolids required by Danish legislation (Nielsen & Bruun 2015; Larsen *et al.* 2017a). Furthermore, it is odourless – a feature that gives it an advantage over mechanically treated and subsequently stored sludge, which has a strong odour, thus making it difficult to aside, even if it meets the threshold values for metals and xenobiotics. Therefore, longer transport distances are often required, as there are fewer land application sites available for receiving dewatered sludge.

Life cycle inventory data on biological gas emissions, substance flows between sub-processes and long-term emissions related to land application were prepared based on data obtained from the data generation activities described in Chapters 2.2, 2.3 and 2.4. Gas emission rates, percentage shares of C and N lost to different gas species during treatment and substance flows are presented in Larsen *et al.* (2017d), sections SI-4 and SI-5, while emissions related to land application are presented in section SI-7.

Based on the mass flow analysis of the different treatment technologies obtained from the activities related to research area 2 (Chapter 2.3), the flow of substances through the three scenarios, based on an input corresponding to the FU chosen for the LCA (1000 kg WW of SAS), was calculated in EASETECH. Table 4 provides an overview of the amount of substances allocated to reject water and the treated sludge by the end of the different treatment processes ("final sludge product"). The reduction of water, DS, VS, TC and TN not accounted for through loss to reject water was due to evaporation/evapotranspiration and mineralisation processes.

Based on the gas emission rates obtained from activities related to research area 1 (Chapter 2.2), the percentage shares of C and N emitted as CH_4, CO_2, N_2, N_2O and NH_3 were calculated for the different treatment technologies (Table 5). The environmental loadings related to gas emissions from mineralisation processes during the different scenarios, calculated in EASETECH, were based on these values.

Emissions of C and N related to land application, covering 100 years, obtained from the activities related to research area 3 (Chapter 2.4) are shown in Tables 6 and 7. Based on these values, environmental loadings related to land application were calculated in EASETECH.

Table 4. Allocation of substances to reject water and final sludge products based on an input of 1000 kg WW of SAS.

Input			Reject water		Final sludge product for land application		
1000 kg of SAS			Centrifuge	STRB system	S-CEN	S-STRB	S-SPA
Wet Weight	kg	1000.00	974.00	955.32	25.53	11.55	8.60
Water	kg	993.21	973.35	954.99	19.52	8.84	6.18
DS	kg	6.79	0.65	0.34	6.01	2.72	2.42
VS	kg	4.17	0.30	0.16	3.73	1.72	1.32
TC	kg	1.89	0.14	0.10	1.64	0.76	0.56
TN	g	269.56	18.97	25.70	247.52	106.79	93.85
P	g	155.49	9.92	0.96	145.57	154.54	154.54
K	g	26.56	2.90	1.57	23.65	24.95	24.95
NH4+-N	mg	1.03E-01	6.56E-03	0.00E+00	2.49E-01	0.00E+00	0.00E+00
NO3--N	mg	6.79E-06	7.43E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Mg	g	28.75	7.89	1.38	20.85	27.18	27.18
Ca	g	191.85	49.51	12.05	142.35	179.57	179.57
Cr	g	1.56E-01	1.61E-02	3.33E-03	1.40E-01	1.53E-01	1.53E-01
Mn	g	5.07	0.52	0.26	4.55	4.83	4.83
Fe	g	434.36	33.26	17.09	401.10	418.02	418.02
Ni	g	0.15	0.01	0.00	0.14	0.15	0.15
Cu	g	2.13	0.08	0.02	2.05	2.12	2.12
Zn	g	3.89	0.15	0.09	3.73	3.78	3.78
Cd	g	4.45E-03	2.04E-04	7.05E-05	4.25E-03	4.38E-03	4.38E-03
Pb	g	2.02E-01	1.22E-02	3.94E-04	1.90E-01	2.01E-01	2.01E-01

Table 5. Average, daily rates and percentage shares of C emitted as CO_2 and CH_4 and N emitted as N_2, N_2O and NH_3 from 1) stored, centrifuged sludge, 2) sludge residue during treatment in an STRB system and 3) sludge residue during post-treatment at an SPA.

	1) Storage subsequent to centrifuging				2) Treatment in STRB system		3) Post-treatment at SPA	
	On-site storage - 1 week		External storage - 1 year		12 years		4 months	
	$mg \cdot m^{-3} \cdot day^{-1}$	% of C	$mg \cdot m^{-3} \cdot day^{-1}$	% of C	$mg \cdot m^{-3} \cdot day^{-1}$	% of C	$mg \cdot m^{-3} \cdot day^{-1}$	% of C
CO_2-C	56,344	53%	8,128	48%	21,257	93%	47,406	92%
CH_4-C	49,881	47%	8,935	52%	1,597	7%	3,932	8%
	$mg \cdot m^{-3} \cdot day^{-1}$	% of N	$mg \cdot m^{-3} \cdot day^{-1}$	% of N	$mg \cdot m^{-3} \cdot day^{-1}$	% of N	$mg \cdot m^{-3} \cdot day^{-1}$	% of N
N_2-N	1,965	67%	982	75%	2,926	95%	14,980	98%
N_2O-N	964.65	33%	267	25%	7	5%	272	2%
NH_3-N	11.3	0%	2	0%	0	0%	0	0%

Table 6. Emission factors of nitrogen after application of sludge residues on a sandy loam soil under an average Danish precipitation regime, compared with the application of mineral fertiliser.

Sludge type	Per	NH_3-N	N_2O-N	NO_3^--N (groundwater)	NO_3^--N (surface)	N crop uptake
Centrifuge-separated SAS	% input N	2.3%	2.8%	32.5%	12.1%	8.4%
STRB system immediate application	% input N	2.10%	3.00%	27.70%	10.30%	6.10%
STRB system with solar drying	% input N	0.7%	3.1%	31.0%	11.4%	7.5%

Table 7. Fraction of the initial application of 30 kg P ha^{-1} in sludge residues, which resulted in the lower application of mineral P fertiliser (substitution, F_{PFS}) and increased P loss (F_{Ploss}) over the subsequent 100 years.

Sludge r type	Per	Carbon sequestration	Carbon dioxide emission
Centrifuge-separated SAS	% input C	12.5%	87.5%
STRB system immediate application	% input C	13.5%	86.5%
STRB system followed by post-treatment at SPA	% input C	19.0%	81.0%

Impact Assessment

Fourteen mid-point impact categories were included in the LCA (Table 8).

Table 8. Impact categories and normalisation factors used in the LCA.

Impact category	LCIA method	Unit	Normalization reference
Climate Change	IPCC 2007	kg CO_2-eq	$8.1 \cdot 10^{3}$
Stratospheric Ozone Depletion	EDIP	kg CFC-11-eq	$4.1 \cdot 10^{-2}$
Human Toxicity, Carcinogenic	USEtox	CTU	$5.4 \cdot 10^{-5}$
Human Toxicity, Non-Carcinogenic	USEtox	CTU	$1.0 \cdot 10^{-4}$
Ionising Radiation	ReCiPe Midpoint (H)	kg U235-eq	$1.3 \cdot 10^{3}$
Photo Oxidant Formation	ReCiPe Midpoint (H)	kg NMVOC	$5.7 \cdot 10^{1}$
Freshwater Eutrophication	ReCiPe Midpoint (H)	kg P-eq	$6.2 \cdot 10^{-1}$
Marine Eutrophication	ReCiPe Midpoint (H)	kg N-eq	9.3
Ecotoxicity	USEtox	CTU	$6.7 \cdot 10^{2}$
Depletion of Fossil Abiotic Resources	CML 2012	MJ	$6.2 \cdot 10^{-4}$
Depletion of Reserve-Based Abiotic Resources	CML 2013,	kg Sb-eq	$3.4 \cdot 10^{-2}$
Terrestrial Acidification	Accumulated Exceedance	AE	$5.0 \cdot 10^{1}$
Terrestrial Eutrophication	Accumulated Exceedance	AE	$1.2 \cdot 10^{2}$
Particulate Matter	updated from ILCD (2010)	Kg PM2.5-eq	2.8

The choice of impact categories and LCIAs for the impact categories was made according to recommendations stated by the Institute for Environment and Sustainability in the European Commission Joint Research Centre (JRC) (ILCD 2010). Environmental impacts caused by the different impact categories were normalised according to normalisation factors found in Blok *et al.* (2013), so the units for very impact category were converted into people equivalents (PE), representing the annual impact of an average person in relation to the various impact categories.

Sensitivity analysis

To test the robustness of the results of the LCA, a two-step robustness analysis was performed. The first step was to perform a contribution analysis identifying those substances influencing more than 90% of the overall environmental impact of the different impact categories. Second, a sensitivity analysis (SA) was conducted to test how the results of the LCA were affected by changes in specific parameters in the scenarios. When running the analysis, specific parameters were tested separately, ensuring that changes related to the specific parameters do not affect each other. The parameters chosen for the SA were mineralisation rates and transport distances. In SA-1, it was tested how increasing or decreasing the amounts of C and N mineralised during treatment in an STRB system or an SPA, or storage subsequent to mechanical dewatering by 10% of its original value, affected the outcome of the LCA. In SA-2, it was tested how changing the transport distances affected the outcomes of the LCA. First, transport distances included in the scenarios were reduced by 50%. Second, transport distances included in S-STRB and S-SPA were left unchanged, while those included in S-CEN were reduced from 70 and 200 km to 0.150 and 10 km, respectively, now being the same as the distances included in S-SPA.

4 Results

In the first part of this chapter, the main results and conclusions of the data generation activities related to the STRB system technology are presented, while last part presents and discuss the results of the LCA. The data generated for the mechanical treatment technology are presented in Larsen *et al*. (2017d).

4.1 Research area 1: Quantification of biological gas emissions from sludge treatment

The results for seasonal gas emissions from STRB systems and changes in gas composition in the pore space of sludge residue residing in a bed related to loading state are presented in Larsen *et al*. (2017c) and Larsen *et al.* (2017b).

The average daily emission rates of CO_2, CH_4 and N_2O, based on measurements carried out over four seasons at Helsinge's STRB system, are shown in Figure 11. These emission rates varied significantly over the year. For CO_2 and CH_4, rates recorded during summer were the highest of all seasons, while the lowest rates were recorded during winter. For CO_2, emission rates measured during spring and autumn were very similar. For CH_4, the rate measured during spring was the second highest, and it was twice the rate recorded in autumn. For N_2O, the highest emission rate was recorded during autumn and the lowest during spring, while emissions recorded during summer and winter were similar and found in between those of autumn and spring.

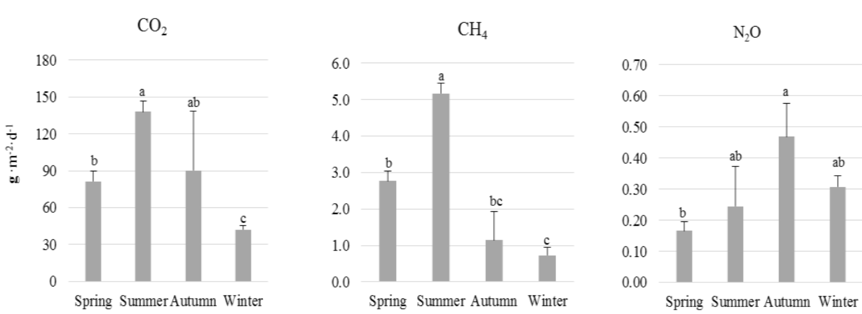

Figure 11: Seasonal emission rates of CO_2, CH_4 and N_2O from sludge residue in an STRB system. Bars indicate standard errors. Letters indicate significant differences among the seasons for each gas species; the letters cannot be compared among the different gas species.

Microbial activity is lower during colder seasons, resulting in slower gas emission rates and a build-up of organic material. When the temperature increases during spring, microbial activity thus increases, resulting in increasing gas emission rates due to degradation of the accumulated organic material (Vincent *et al.* 2011). This figure was applied to the seasonal changes in emissions observed for CO_2 and CH_4, though not for N_2O. Many factors influence the generation of N_2O, which can be produced under both anaerobic and aerobic conditions (Lloyd *et al.* 1987; Robertson *et al.* 1995; Gui *et al.* 2007; Zhou *et al.* 2008; Kampschreur *et al.* 2009). Firestone *et al.* (1980) found that N_2O is produced primarily in substrates during the transition from anaerobic to aerobic conditions. Indeed, for our results, there was a tendency that the emission of N_2O increased after loading, peaked and then declined to a steady state (Larsen *et al.* 2017c, b).

When a bed in an STRB system is loaded with sludge, the pore space becomes water-locked (Vincent *et al.* 2012); however, this water starts to drain from the sludge residue immediately after sludge loading, and 24 hours thereafter approximately 96% of the water contained by the sludge has drained off. During the subsequent resting period, more water leaves the sludge residue, due to draining and evapotranspiration, thus allowing O_2 to re-enter the sludge residue gradually (Nielsen 1993; Vincent *et al.* 2012). However, data on gas composition in the pore space of the sludge residue in STRB systems are scarce. Therefore, as an additional experiment, percentage gas compositions at various depths in the sludge residue at Helsinge's STRB system during a resting period were recorded (Larsen *et al.* 2017b), alongside surface emissions of CO_2, CH_4 and N_2O. During this experiment, the surface emissions of N_2O increased steadily during the resting period. Furthermore, the percentage content of O_2 in the pore space of the sludge residue also rose during this period, suggesting that the production of N_2O happens during the transition from anaerobic to aerobic conditions. However, as N_2O is produced as a by-product of denitrification, the emission rate of N_2O also depends on the amount of NO_3^- available to the denitrifying bacteria. The amount of NO_3^- present in the sludge residue is highest immediately after sludge application, which, together with the transition from anaerobic to aerobic conditions, causes the production of N_2O to rise. These dynamics could explain why the emissions of N_2O presented in Larsen *et al.* (2017c) peaked during autumn: heavy rainfall was recorded during this season, meaning that the sludge residue was soaked during the resting period. Therefore, it was constantly in a transitional state between aerobic and anaerobic conditions, resulting in high emission rates of N_2O.

Gas emission dynamics are of interest in relation to the climatic impact of the sludge treatment process in STRB systems. Emissions of CO_2 related to the treatment of wastewater and sludge are considered climate-neutral (IPCC 2007). Nonetheless, CH_4 and N_2O are potent greenhouse gasses, with GWPs of 28 (excl. carbon feedbacks) and 265 (excl. carbon feedbacks), respectively. The results of the seasonal gas emission recordings revealed that the emission rates of CH_4 and N_2O change considerably during seasons. Therefore, seasonal variations should be taken into account when calculating the annual GWP for gas emissions originating directly from the mineralisation process. As N_2O has a high GWP, even small changes in the annual emission rate will cause considerable changes for the total GWP of the treatment process. It is therefore of interest to operate STRB systems in a way that minimises the emissions of these gas species.

4.2 Research area 2: Substance flows in an STRB system and an SPA

The results on substance flows in an STRB system and an SPA are presented in Larsen *et al.* (2017a).

Samples of sludge, reject water and sludge residue of different ages were analysed for contents of DS, VS, TC, TN, NO_3^-, NH_4^+, P, K, Ca, Fe, Mn, Cr, Ni, Cu, Zn, Cd and Pb. Additionally, a substance flow analysis, covering 12 years of treatment in an STRB system and three months of post-treatment at an SPA, was performed. For the STRB system, substance flows were divided into five streams, namely loss to reject water, loss to mineralisation, accumulation in sludge residue and evapotranspiration (only relevant for water) (Table 9a). For the SPA, substance flows were divided into four streams, namely loss to reject water, loss to mineralisation, accumulation in sludge residue and evapotranspiration (Table 9b).

Over the 12 years of treatment, contents of VS, TC and TN in sludge loaded into an STRB system reduced by more than 50% (Table 9a). Mineralisation rates during the first year of treatment were the highest, with the rates for VS, TC and TN being 57%, 54% and 52%, respectively, while the average annual rates for the subsequent 11 years were 3%, 3% and 5%, respectively. These changes in mineralisation rates reflect that the main share of easily degradable organic matter provided by the sludge is mineralised during the first year of treatment, albeit mineralisation activity continues during the entire treatment period, admittedly at a lower rate.

Reeds growing in the sludge residue of an STRB system are never harvested but wither due to their natural life cycle, the dead reeds being incorporated into the sludge residue and mineralised by the same microbial processes. As reeds extract C from the atmosphere through photosynthesis, dead reeds act as an extra source of C to the system. However, as the amount of C supplied by the sludge is much greater compared to the amount fixated by the reed, the input of C from the reeds was assumed negligible in the C budget. Reeds also take up a share of the N, P and metals contained in the sludge residue, so a share of these substances cycles continually between the sludge residue and the standing reed.

During three months of post-treatment at the SPA, the mineralisation rate increases, whereas the contents of VS, TC and TN in the sludge residue were reduced by 25, 25 and 12%, respectively. When sludge residue is excavated from an STRB system, it is mixed and aerated due to the excavation activity, thereby enhancing aerobic mineralisation. The greenhouse roof and walls covering the storage area add a solar drying effect to the treatment, further enhancing microbial activity and evaporation. Furthermore, the reeds, which are incorporated into the sludge residue due to the excavation activity, add fresh, organic material to be mineralised. The C accumulated in the reeds originates from the atmosphere and therefore does not count in the C budget. Moreover, the addition of fresh organic material to the sludge residue stimulates microbial activity; nonetheless, N contained by the reeds counts in the N budget, and so post-treatment at an SPA allows for a share of the N held in the standing reeds to be mineralised.

The effective mineralisation of C and N during treatment in STRB systems and SPAs results in a well-stabilised final sludge product. This is noteworthy, since the common fate of sludge residue is land application, and less stabilised organic material has a greater potential for causing eutrophication and emitting greenhouse gasses (Yoshida *et al.* 2015; Gómez-Muñoz *et al.* 2017). The more stabilised the final sludge product, the fewer environmental effects due to N_2O and NH_3 emissions and leaching of NO_3^- to ground- and surface water.

For P, K and metals, the only way to leave the system is via reject water. Therefore, the major shares of most metals (> 90%) are accumulated in the final sludge residue. However, the share leaching into reject water never truly leaves the system, since the latter is returned to the WWTP, where it is mixed with incoming wastewater and treated again. The resulting SAS is treated via exactly the same sludge treatment procedure, meaning that the major part of the non-degradable substances eventually will end up in sludge residue (a minor share is lost to reject water leaching from the wastewater treatment process and into the environment).

Table 9 The distribution (%) of substances in feed sludge after treatment in an STBR system and after post-treatment at an SPA. Substance concentrations in the accumulated sludge residue and in the sludge residue after post-treatment are also given. For comparison purposes, threshold values of heavy metals, as stated by the Danish Environmental Protection Agency and the European Union, are shown.

a: Substance flow in STRB system (during 12 years)																	
	DW	WATER	VS	C	N	P	K	Mg	Ca	Cr	Mn	Ni	Fe	Cu	Zn	Cd	Pb
Streams:	100%	100%	99%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Loss to reject water	4%	96%	5%	5%	9%	1%	4%	1%	12%	0%	5%	4%	0%	1%	1%	1%	0%
Loss to evapotranspiration, during 12 years of treatment	-	3%	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Loss to mineralisation, during 12 years of treatment	56%	-	55%	56%	51%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Accumulation in sludge residue, during 12 years of treatment	40%	1%	39%	40%	40%	99%	96%	99%	87%	100%	95%	96%	100%	99%	99%	99%	100%
Average conc. in accumulated sludge residue (mg ·kg-1 DM)	-	-	-	-	-	-	-	-	-	35	-	25.26	-	317	1099	1.26	42
Average conc. in accumulated sludge residue (mg ·kg-1 TP)	-	-	-	-	-	-	-	-	-	-	-	568.65	-	-	-	28.27	946.96
b: Substance flows at SPA (3 months of treatment)																	
	DW	WATER	LOI	C	N	P	K	Mg	Ca	Cr	Mn	Ni	Fe	Cu	Zn	Cd	Pb
Streams:	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Loss to evaporation	-	30%	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Loss to mineralisation	13%	-	25%	26%	12%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Accumulation in sludge residuetreatment	87%	70%	75%	74%	88%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Average conc. in accumulated sludge residue (mg ·kg-1 DM)	-	-	-	-	-	-	-	-	-	0.03	-	0.03	-	362	699	1.01	35
Average conc. in accumulated sludge residue (mg ·kg-1 TP)	-	-	-	-	-	-	-	-	-	-	-	1.04	-	-	-	35.22	1218.03
c: Treshhold values for heavy metals in bio solids intended for land application																	
Denmark																	
BEK No. 1650 of 13/12/2006 (mg ·kg-1 DM)	-	-	-	-	-	-	-	-	-	100	-	30	-	1000	4000	0.8	120
BEK No. 1650 of 13/12/2006 (mg ·kg-1 TP)	-	-	-	-	-	-	-	-	-	-	-	2500	-	-	-	100	10000
EU																	
86/278/EEC EU Directive (mg ·kg-1 DM)	-	-	-	-	-	-	-	-	-	-	-	300-400	-	1000-1750	2500-4000	20-40	750-1500
ENV. E 3 (2000) Working document on sludge., 3rd draft (mg ·kg-1 DM)	-	-	-	-	-	-	-	-	-	1000	-	300	-	1000	2500	10	750

Consequently, the amount of P, K and metals applied to the land comes down to the initial content of wastewater treated by the WWTP. However, the threshold concentrations of heavy metals for biosolids intended for land application are stated by Danish legislation (BEK No. 1650 of 13 December 2006) (Table 9c).

If treatment in an STRB system is combined with post-treatment at an SPA (as in S-SPA), the accumulation of organic matter, TC and TN in the final sludge product is 30 to 35% of the amounts initially supplied by the sludge, while the overall reduction, calculated as total wet weight, for the entire treatment process is 99%.

4.3 Research area 3: Emissions associated with sludge applied to the land

The results for emissions associated with land application are presented in Gómez-Muñoz *et al.* (2017) and Larsen *et al.* (2017d).

To investigate the dynamics of CO_2 and N_2O emissions and NO_3^- and NH_4^+ leachate from sludge treated in STRB systems, samples representing the different technologies were incubated over 160 days under conditions simulating application to agricultural land in Denmark. During incubation, emissions of CO_2 and N_2O and leaching of NO_3^- and NH_4^+ were continually recorded.

Figures 12 and 13 show the recorded emissions of CO_2 and N_2O and the leaching of NO_3^- and NH_4^+. The results reveal that the emission rates of CO_2 and N_2O from older sludge residue stabilised more quickly compared to younger sludge residue (Figure 12a), suggesting that more of the readily degradable organic matter in sludge residue subjected to longer treatment was mineralised (which is consistent with the results on mineralisation rates presented in Chapter 4.2).

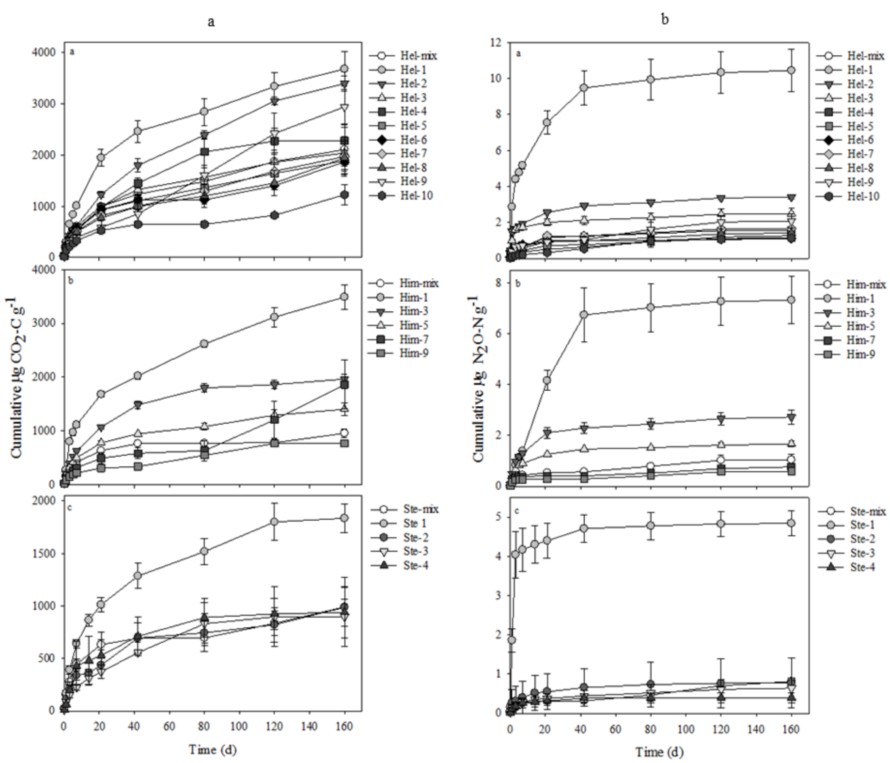

Figure 13: Emission rates of CO_2 (a) and N_2O (b) from sludge residue of different ages and origin when applied to the land. The samples were incubated for 160 days under conditions simulating land application in Denmark. The sample IDs refer to the origin and age of the samples: Hel: Helsinge STRB system (Denmark). Him: Himmark STRB system (Denmark). Ste: Stenlille STRB system (Denmark). 1: Oneyear of treatment. 2: Two years of treatment etc. Mix: Composite sample based on samples of sludge residue subjected to different treatment times (Helsinge 1 – 10 years; Himmark 1 – 9 years; Stenlille 1 – 4 years).

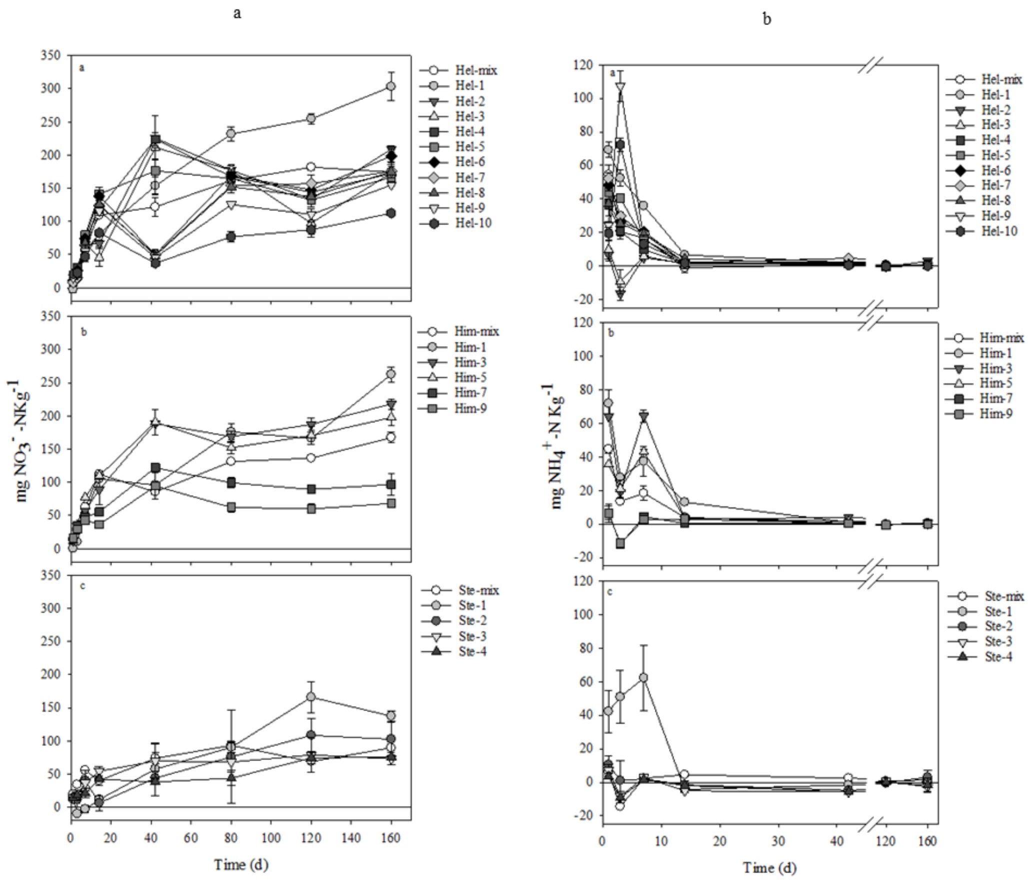

Figure 13: Accumulation of NO_3^- (a) and NH_4^+ (b) in sludge residue of different ages and origin when applied to the land. The samples were incubated for 160 days under conditions simulating land application in Denmark. The sample IDs refer to origin and age of the samples: Hel: Helsinge STRB (Denmark). Him: Himmark STRB(Denmark). Ste: Stenlille STRB (Denmark). 1: One year of treatment. 2: Two years of treatment etc. Mix: Composite sample based on samples of sludge residue subjected to different treatment times (Helsinge 1 – 10 years; Himmark 1 – 9 years; Stenlille 1 – 4 years).

For all samples, the concentration of NH_4^+ decreased to zero within the first 15 days of the experiment.

For some of the samples, unrelated to age, concentration rose during the very first days and then declined. With respect to NO_3^-, concentration in the younger samples steadily rose during the incubation period (Figure 13), while the older samples tended to stabilise, suggesting that NH_4^+ available for nitrification depleted faster in the older samples. The composite samples, representing the actual composition of sludge residue used for land application, were among the most stabilised samples, suggesting that even though they contain sludge residue subjected to shorter treatment periods, they have low eutrophication potential.

4.4 Results of the LCA

The loadings and savings calculated for each impact category were sorted into six sub-categories:

Daily operation: Electricity consumption for the daily pumping of sludge and reject water. In S-CEN, this sub-category also includes polymer coagulant consumption.

Biological gas emissions: Gas emissions related to mineralisation processes during treatment in the STRB system, post-treatment at the SPA or storage subsequent to dewatering on a centrifuge.

Transport/excavation: Fuel consumption for transport and excavation activities. Also includes application to the land by a tractor.

Land application: Gaseous emissions, leaching of substances and carbon sequestering related to land application of treated sludge.

Fertiliser substitution: The effect of substituting commercial fertiliser by applying treated sludge to the land.

Reject water treatment: Electricity consumption related to WW treatment, gaseous emissions and leaching related to WW treatment, all emissions caused by the re-running of SAS produced through the entire sludge treatment process, including land application and fertiliser substitution.

For all impact categories, a contribution analysis was undertaken to identify the substances causing > 90% of the environmental loadings; the results are to be found in Table 10.

Results of non-toxic environmental impact categories

Climate Change

For the impact category “Climate Change”, all scenarios provided a net-loading (Figure 14). For all scenarios, the major part of the loading was caused by the sub-category “Biological gas emissions”, due to emissions of CH_4 and N_2O (Table 10).

Table 10: List of substances contributing by > 90% of the environmental loadings in the various impact categories and scenarios.

Impact Category	S-CEN	S-STRB	S-SPA
Global warming	CH_4, N_2O	CH_4, N2O	CH_4, N_2O
Freshwater eutrophication	P, PO_3^{4-}	PO_3^{4-}, P	PO_3^{4-}, P
Marine eutrophication	NO_3^-	NO_3^-	NO_3^-
Terrestrial acidification	NH_3	NH_3	NH_3
Terrestrial eutrophication	NH_3	NH_3	NH_3
Human toxicity – non-carcinogenic	Zn	Zn	Zn
Ecotoxicity	Zn, Cu	Zn, Cu	Zn, Cu
Human toxicity – carcinogenic	Ni	Ni	Ni
Depletion of fossil abiotic resources	Hard Coal, Crude oil	Hard Coal	Hard Coal
Depletion of reserve abiotic resource	In, Cd	In, Cd	In, Cd
Particulate matter	NH_3, SO_2	NH_3, SO_2	NH_3, SO_2
Photochemical oxidant formation	NO_x , NMVOC	NO_x, SO_2	NO_x, SO_2
Stratospheric ozone depletion	CFC-11, CFC-13, HCFC-12	CFC-11	CFC-11
Ionising radiation	C-14, Ce-137, I-129, Ra-222, Co-66	C-14, Ce-137, I-129, Ra-222, Co-66	C-14, Ce-137, I-129, Ra-222, Co-66

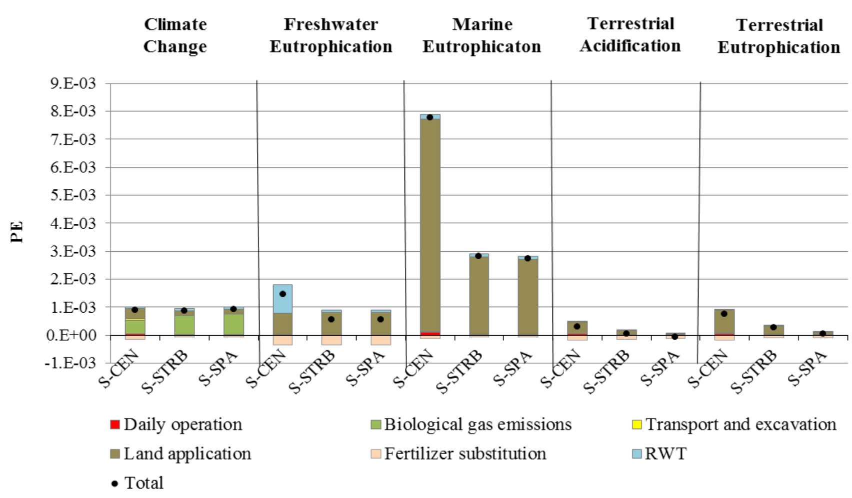

Figure 14: The environmental loadings and savings provided by treatment of 1000 kg ww sludge for the following three treatment scenarios; S-CEN, S-STRB and S-SPA in relation to five non-toxic impact categories. The loadings and savings provided by the different impact categories were converted into people eqvivalents (PE), representing the annual impact of an average person.

The total climate change impacts caused by biological gas emissions during treatment in an STRB system in S-STRB and S-SPA, and storage of the centrifuged sludge in S-CEN, were almost the same. During the 12-year treatment process in the STRB system, the main share of the mineralised C and N was emitted as CO_2 (93%) and N_2 (94%) (Table 5). These gas emissions are climate-neutral, as N_2 is not a greenhouse gas and CO_2 originating from biological sources, such as wastewater and sludge, is considered short-cycled C (IPCC 2007). The remaining C and N were emitted as CH_4 (7%) and N_2O (6%), which have GWPs of 28 (excl. carbon feedbacks) and 265 CO_2 equivalents (excl. carbon feedbacks) (IPCC 2014), respectively. During the six months the mechanically dewatered sludge was stored at the external storage facility, only 48% of the C mineralised was emitted as CO_2, and only 74% of the N emitted as N_2, suggesting that stored dewatered sludge was dominated by anaerobic conditions. The larger share of C and N emitted as CO_2 and N_2 in the STRB system was due to the more efficient aeration of the sludge residue (see description in Chapter 2.2). Hence, a larger share of the C and N mineralised in S-CEN counts as loading in relation to climate change. However, as the *amounts* of C and N mineralised during treatment in the STRB system were much higher compared to amounts mineralised during dewatered sludge storage, the loadings provided by biological gas emissions on climate change were the same for all three scenarios. The lower mineralisation rate during the sludge treatment process in S-CEN means that more of the C and N was found in the final sludge product, which eventually would be applied to the land. Indeed, emissions of N_2O from the land application of sludge residue were higher for S-CEN compared to S-STRB and S-SPA (Table 6). The share of N emitted as N_2O after soil application was approximately 3% for all three sludge products; however, the final sludge product produced in S-CEN contained more N compared to the final sludge products produced from S-STRB and S-SPA (Table 4), thereby resulting in greater loading from S-CEN. For all scenarios, small environmental savings were obtained by substituting mineral fertiliser due to avoided greenhouse gas emissions related to production of commercial fertilizer.

Marine Eutrophication

The impact category "Marine Eutrophication" (Figure 14), for all scenarios, was affected mainly by the leaching of NO_3^- related to land application of the final sludge product (Table 10). The loading provided from land application by S-CEN was much higher compared to the other scenarios. The high loading provided by S-CEN was due to the large amount of N contained by the final sludge residue produced from this scenario (Table 4). Even though the percentage shares of N leaching into surface water from the final sludge product when applied to the land were quiet similar among the scenarios (Table 6), the large amount of N contained by the final sludge product produced by S-CEN led to a greater amount of leaching.

Loadings from all other sub-categories apart from “Land application” were negligible. However, as the demand for fuel and electricity was higher in S-CEN, the loadings provided by “Daily operation” and “Transport and Excavation” in this scenario exceeded the corresponding loading in S-STRB and S-SPA.

Terrestrial Acidification and Eutrophication

The impact categories “Terrestrial Acidification” and “Terrestrial Eutrophication” (Figure 14) were both affected mainly by gaseous emissions of NH_3 (Table 10). However, NH_3 emissions related to the treatment processes for all scenarios were small; indeed, for S-STRB and S-SPA, mineralisation during treatment produced no NH_3 (Table 5). Therefore, the loadings rose mainly from land application. As for “Marine Eutrophication”, the environmental loading caused by S-CEN was twice as high compared to the other scenarios, due to the higher content of N in the final sludge product when applied to the land. However, the magnitudes of the impacts on “Terrestrial Acidification” and “Terrestrial Eutrophication” for all scenarios were much lower compared to “Marine Eutrophication”.

Freshwater Eutrophication

The impact category “Freshwater Eutrophication” (Figure 14) was affected by the amount of P entering the environment. For S-STRB and S-SPA, the major share of the loadings was provided by the leaching of PO_3^{4-} due to land application, while the major loading for S-CEN was provided by PO_3^{4-} leaching from reject water treatment (Table 10). As the reject water produced by the centrifuging process contained 10 times more P compared to the reject water draining from STRB system, more PO_3^{4-} leached into the environment, due to the reject water treatment in S-CEN. Most of the P contained by the final sludge product binds within the soil or is taken up by the crop; hence, the leaching of PO_3^{-} from the final sludge product applied to the land is very low.

Results for toxic impact categories

Figure 15a show the loadings and savings provided for three toxic impact categories. The magnitudes of “Human Toxicity – non-carcinogenic” and “Ecotoxicity” were much higher compared to “Human toxicity – cancer”, so the latter is presented isolated in Figure 15b. All three impact categories were affected mainly by heavy metals (Table 8), and as metals do not degrade, they end up in the reject water or the final sludge product.

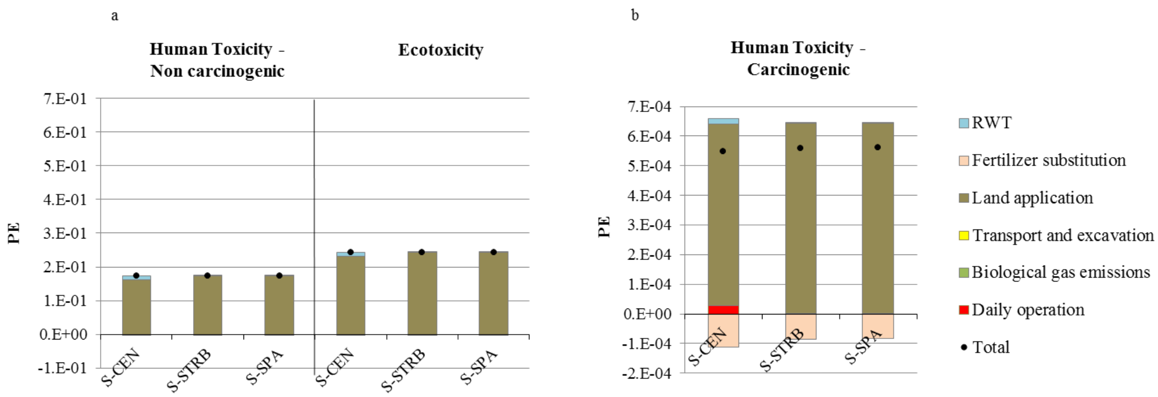

Figure 15: The environmental loadings and savings provided by treatment of 1000 kg ww sludge for the scenarios S-CEN, S-STRB and S-SPA in relation to three toxic impact categories a) The impact categories Human Toxicity – Non carcinogenic and Ecotoxicityb) The impact category “Human Toxicity – Carcinogenic” is presented on a separate y-axis. The loadings and savings provided by the different impact categories were converted into people eqvivalents (PE), representing the annual impact of an average person.

Human toxicity – non-carcinogenic and Ecotoxicity

Loadings affecting “Human toxicity – non-carcinogenic” and “Ecotoxicity” were primarily provided by “Land application”. These impact categories were affected by Zn and Cu, which originates primarily from wastewater treated by sludge treatment lines. Zink and Cu also enter the environment through the combustion of fuel etc.; however, the contributions made by “Daily Operation” and “Transport/Excavation” were negligible compared to the input from the SAS.

For S-CEN, the environmental loading provided by “Land application” was slightly lower compared to S-STRB and S-SPA, as a larger share of the input Zn and Cu was allocated to reject water in S-CEN (Table 4).

However, all scenarios include environmental loadings arising from the re-treatment of reject water at the WWTP and the subsequent re-running of the SAS produced through the entire sludge treatment line, terminated by land application. Hence, almost 100% (except for the very small share leaving the WWTP through the outlet from the WW treating process) of Zn and CU, and all other metals, eventually ends up on agricultural land. Indeed, the loading provided by "Land application" was slightly larger for S-STRB and S-CEN, though the loading provided by "Reject water treatment" in S-CEN was correspondingly larger, as more metals re-entered the WWTP through the reject water in this scenario. As a result, the metals ended up on agricultural land after the second treatment.

Only very small savings were provided by "Fertiliser substitution" for these Impact categories. Zinc and Cu are among the metals essential to plant growth, albeit in small doses. As the substitution of commercial fertilisers means that environmental loadings related to the production of commercial fertiliser were saved, the savings from "Fertiliser substitution" were related to the amount of Zn and Cu avoided from being released into the environment during the production of fertiliser. However, as the amounts of these substances originating from fuel consumption etc. were small compared to the amounts provided by the SAS, the contributions made by "Fertiliser substitution", "Daily operation" and "Transport and Excavation" were negligible.

Human Toxicity – Carcinogenic

The impact category "Human Toxicity – Carcinogenic" (Figure 15b) was mainly affected by Ni applied to land (Table 10). The total impacts provided by the three scenarios were almost the same; however, the sources of the loadings were slightly different. For S-CEN, the major contribution arouses from "Land application", but "Reject water treatment" and "Daily operation" also contribute. For S-STRB and S-SPA, the major loadings also arose from "Land application", though the loadings provided by "Reject water treatment" and "Daily operation" are negligible. Differences in the sources of loadings were due to the same circumstances as explained for "Human toxicity – non-carcinogenic" and "Ecotoxicity", in that the loading provided by "Daily operation" in S-CEN was related to the production of the polymer coagulant required in this scenario. The environmental impacts caused by Ni originating from the production of commercial fertiliser were greater compared to the impacts caused by Zn and Cu of the same origin; hence, savings made from avoiding the production of commercial fertiliser were larger in "Human Toxicity – Carcinogenic" compared to the other two toxic impact categories. As the final sludge product produced from S-CEN contains much more N compared to the final sludge product produced from the other scenarios, it supressed more production of commercial fertiliser, and thereby provided a greater saving. However, as the loadings provided by the other life stages in S-CEN were higher compared to S-STRB and S-CEN, the overall result was identical impacts in all scenarios.

Results for impact categories related to resource consumption

Depletion of fossil abiotic resources

For S-CEN (Figure 16), the loadings were provided by the consumption of hard coal and crude oil (Table 10), while S-TRB and S-SPA only consumed hard coal. As the consumption of crude oil in S-Cen was related to the production of the polymer coagulant, the contribution was included in "Daily operation". For S-STRB and S-SPA, only electricity for pumping activity was needed for daily operations, meaning that the impacts from this sub-category were smaller for these scenarios compared to S-CEN.

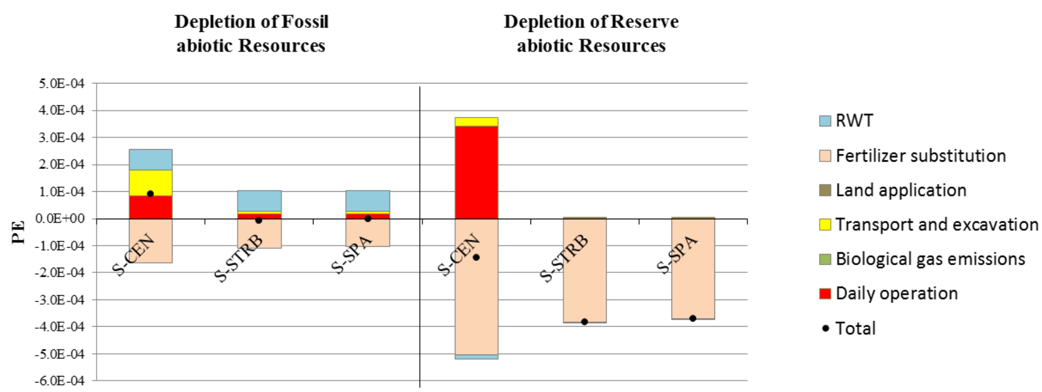

Figure 16: The environmental loadings and savings provided by treatment of 1000 kg ww sludge for the scenarios S-CEN, S-STRB and S-SPA for two impact categories related to resource depletion. The loadings and savings provided by the different impact categories were converted into people eqvivalents (PE), representing the annual impact of an average person.

As S-CEN included much longer transport routes (40 km to external storage and 200 km to the land application site) compared to the other scenarios (0.150 km from the STRB system to the SPA and 10 km to the land application site), fuel consumption related to "Transport/Excavation" was also considerably higher for S-CEN.

The amount of fossil resources saved by avoiding the production of commercial fertiliser, expressed in “Fertiliser Substitution”, was slightly higher for S-CEN, as the final sludge product produced from this scenario substituted more fertiliser due to a higher content of N. However, for S-STRB and S-SPA, the savings caused by fertiliser substitution were greater than the loadings caused by the other sub-categories, resulting in an overall environmental saving for both scenarios. The overall impact from S-CEN was an environmental loading, as the saving caused by fertiliser substitution was not large enough to balance out the loadings caused by daily operations and transport/excavation.

Depletion of reserve abiotic resources

For “Depletion of Reserve abiotic Resources” (Figure 16), loadings arising from “Daily operation” and “Transport/Excavation” in S-STRB and S-SPA were very small. The loading arising from “Daily operation” for S-CEN was large, though, due to a demand for In and Cd (Table 8) related to the production of the polymer coagulant. The production of fuel consumes abiotic resources. As the demand for transport was high in S-CEN, the loading provided by “Transport/Excavation” was correspondingly higher compared to the other scenarios.

“Reject water treatment” also provided a small saving for all scenarios. These savings were related to fertiliser substitution rising from the second run through the sludge treatment line included in “Reject water treatment”. As the final sludge product produced from S-CEN substituted more commercial fertiliser due to a higher N content, the saving made from avoiding the production of commercial fertiliser was higher compared to the other scenarios. For all scenarios, the savings from fertiliser substitution were greater than the loadings provided by the other stage. However, even though the saving for S-CEN was higher compared to S-STRB and S-SPA, resource consumption from the production of the polymer coagulant meant that the overall saving from S-CEN was smaller compared to the other scenarios.

Results for impact categories related to atmospheric pollution

The loadings and savings of four impact categories related to atmospheric pollution are shown in Figure 17.

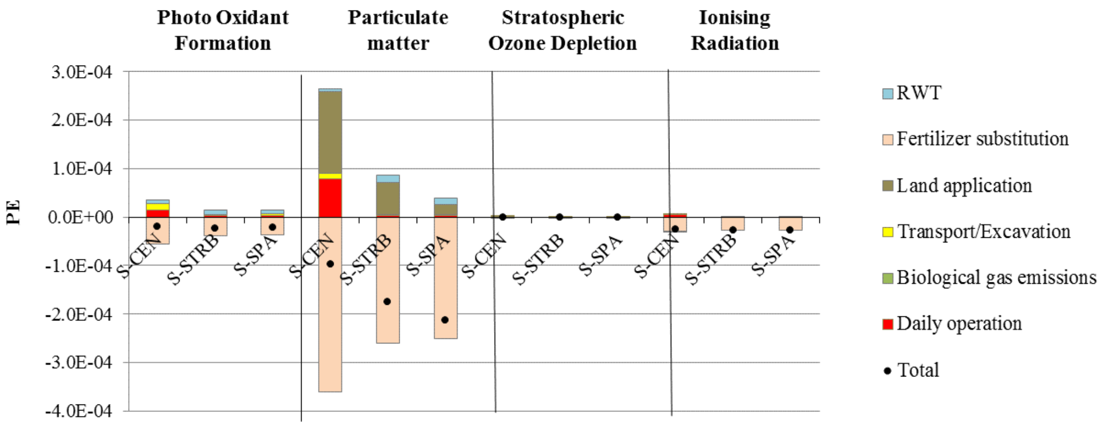

Figure 17: The environmental loadings and savings provided by treatment of 1000 kg ww sludge for the scenarios S-CEN, S-STRB and S-SPA in relation to four impact categories related to atmospheric pollution. The loadings and savings provided by the different impact categories were converted into people eqvivalents (PE), representing the annual impact of an average person.

The impact category "Particulate Matter" is affected by emissions of NH_3 and SO_2 (Table 8). For all scenarios, the largest loadings arose from NH_3 emissions related to land application. As the final sludge product produced by S-CEN contained more N, the impact on "Land application" from this scenario was higher compared to the other scenarios. As NH_3 also arose from fuel consumption, the contributions made by "Daily operation" were high in S-CEN, due to the consumption of fuel related to the production of the polymer coagulant. As the reject water produced and re-treated in S-STRB and S-SPA contained more N compared to S-CEN (Table 7), the loading from "Reject water treatment" was slightly greater for S-STRB and S-SPA. For all scenarios, the savings caused by avoiding the production of commercial fertiliser exceeded the loadings provided by the other stages. Overall, the loading from S-CEN was greater compared to the other scenarios.

For the last three impact categories, "Photo Oxidant formation", "Stratospheric ozone depletion" and "Ionising radiation", the overall impact for all scenarios were savings caused by fertiliser substitution, albeit very small.

4.5 Sensitivity analysis

To test the outcome of the LCA if introducing various changes to the default scenarios, a sensitivity analysis (SA) was carried out. As the interpretation of the results revealed that one of the most crucial parameters affecting environmental impacts was the amount of N contained by the final sludge product, SA-1 tested how raising or lowering the percentage share of N mineralised during treatment or storage affected the outcome of the LCA. Changes in the mineralisation of C were also included. Furthermore, as one of the most striking practical differences between the scenarios was transport distance, SA-2 tested how changing this parameter affected the outcomes of the LCA. SA-1 and SA-2 were carried out separately, meaning that changes made for the mineralisation of C and N, and for transport routes, did not interfere.

All impact factors were included in SA-1 and SA-2. However, not all impact categories were affected by the changes. For SA-1, the most relevant changes in outcomes were seen for "Climate Change" and "Marine Eutrophication", while for SA-2 the most relevant in this regard was "Depletion of fossil abiotic resources". These results are illustrated in Figure 18. The results for SA-1 and SA-2 for all impact categories are shown in Larsen *et al.* (2017d), in section SI-10.

SA-1: Mineralisation rates

In SA-1, the mineralisation rates for C and N were increased and decreased by 10% of their original values in all three treatment scenarios (Figure 18). Mineralisation rates during the treatment of sludge in the STRB system, in the SPA or while storing mechanically dewatered sludge at the external storage facilities affected the emissions CH_4 and N_2O and thereby the impact category "Climate Change". Furthermore, changing the mineralisation rates affected "Marine Eutrophication", as the amount of N found in the final sludge product from the various scenarios depended on the amount of N mineralised earlier in the treatment process.

For "Climate Change", S-CEN was affected more by changes in the mineralisation rates than S-STRB and S-SPA, because a greater share of the C and N mineralised in S-CEN was emitted as CH_4 and N_2O compared to the other two scenarios (Table 5). When the mineralisation rates for C and N were decreased by 10% of their original values for all scenarios, S-CEN provided a lower impact on "Climate Change" than the other scenarios, while it provided a higher impact when the mineralisation rates were increased by 10%.

Due to a higher mineralisation rate in the STRB system while storing centrifuged sludge, S-STRB and S-SPA showed changes in "Marine Eutrophication". However, changing the mineralisation rates for C and N had only a small effect on its impact in the S-CEN.

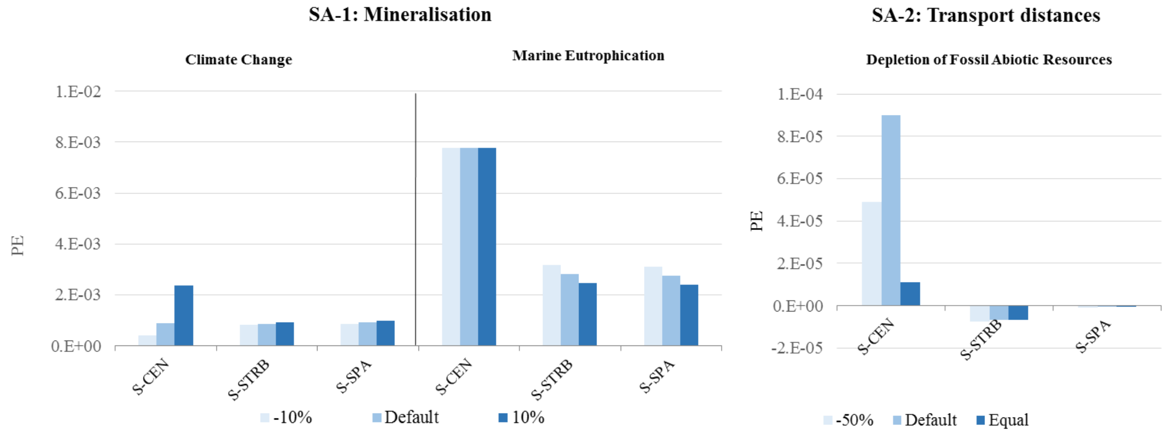

Figure 18. Results of the sensitivity analysis (SA) testing the robustness of the results in relation to mineralisation rate (SA-1) and transport distance (SA-2) in the treatment scenarios S-CEN, S-STRB and S-SPA. "Default" bars represent total impacts caused by the different scenarios in the LCA modelling. For SA-1, "-10%" and "+10%" represent changes in the impact categories "Climate Change" and "Marine Eutrophication" for the different scenarios, if the amounts of mineralised C and N decrease or increase by 10%. For SA-2, "-50%" represents the impacts to "Depletion of Fossil Abiotic Resources", if the transport distances in all scenarios are reduced by 50%. "Equal" represents impacts caused if the transport distances in all scenarios are set to 10 km.

A higher mineralisation rate means that less N remains in the final sludge product, leading to a lower "Marine Eutrophication" impact, while a lower mineralisation rate leads to a higher "Marine Eutrophication" impact. However, regardless of the mineralisation rate applied, the impact on "Marine Eutrophication" in S-CEN was always more than twice as high compared to S-STRB and S-SPA.

The results of SA-1 reflected a trade-off between the impact on "Climate Change" and on "Marine Eutrophication" for the mineralisation rates of C and N during treatment or storage. Higher mineralisation rates led to a higher "Climate Change" impact for S-CEN but a lower "Marine Eutrophication" impact for S-STRB and S-SPA, while lower mineralisation rates had the opposite effect.

SA-2: Transport distances

As S-CEN included longer transport distances compared to S-STRB and S-SPA, this scenario was more affected by changes in transport distances (Figure 18). Reducing the transport distances by 50% reduced the depletion of fossil resources in S-CEN by almost 50%, while this change had no effect on S-STRB and S-SPA. Changing the transport distances included in S-CEN to being the same as in S-SPA, reduced the depletion of fossil fuels to one tenth of the amount depleted in the default S-SPA. However, even if the transport distances included in all three scenarios were the same, S-CEN still depleted more fossil abiotic resources compared to S-STRB and S-SPA. This was due to the demand for the polymer coagulant in S-CEN, the production of which consumes crude oil (Table 10).

4.6 Discussion

What does the presence of an SPA add to STRB systems?

When interpreting the results, focus falls on the differences between S-CEN and the two scenarios representing STRB systems. However, there are also some differences between S-STRB and S-SPA, though these are less pronounced. In most impact categories, total impacts provided by S-STRB and S-SPA were almost identical. However, for the impact categories "Terrestrial Acidification", "Terrestrial Eutrophication" and "Particulate Matter", total impacts caused by S-SPA were lower compared to S-STRB. These impact categories were all affected mainly by emissions of NH_3 originating from land application. The percentage of N in the final sludge product emitted as NH_3 when applied was lower for S-SPA compared to S-CEN (Table 6); furthermore, the amount of N contained by the final sludge residue from S-SPA was reduced by one-third compared to S-STRB, hence the lower emissions of NH_3 from S-SPA.

Despite these differences, the environmental impacts caused by S-STRB and S-SPA were rather similar. Nevertheless, it should be remembered that the environmental impacts evaluated by the 14 impact categories included in the LCA are not the only factors to consider when evaluating whether an SPA is a useful addition to an STRB system. An important consideration, which was not reflected in the LCA, is how the presence of an SPA affects the operational dynamics of the treatment process. As described in section 2.3, the excavation of sludge residue in spring allows the reeds to regrow within the following three to four months, whereas if excavation happens in autumn, the reeds will not regrow until the next coming spring/summer, almost one year later. The shorter re-growth period means that the other beds in the STRB system need to receive an increased amount of sludge for a shorter period, as the loading scheme returns to normal faster. In the longer run, this means it is easier to keep the beds healthy and avoid overloading, and that the treatment capacity is increased without adding more beds to the STRB system.

Comparing STRB system with mechanical treatment

Based on the results of the LCA, STRB systems performed slightly better compared to the conventional dewatering/storage technology.

The results of the LCA revealed that S-STRB and S-SPA for the impact categories affected mainly by resource-consuming processes (the impact categories shown in Figure 16 and 17) performed better compared to S-CEN, as S-CEN requires the input of polymer coagulant, the production of which is rather resource-consuming and includes longer transport distances. However, compared to the non-toxic impact categories presented in Figure 14, the magnitudes of the effects of the impact categories shown in Figures 16 and 17 were low. For all scenarios, the major environmental loading on the non-toxic impact categories was related to biological greenhouse gas emissions from the treatment process and to the amount and fate of N in the final sludge products. Thereby, the factors influencing these impact categories the most for both technologies were whether the mineralisation processes were aerobic or anaerobic. However, if the performances in relation to mineralisation and biological gas emissions were the same for both technologies, STRB systems would perform slightly better compare to the conventional technology due to the small, but present, differences in resource consumption.

Even though the environmental performance of a sludge treatment technology is important when deciding which one to implement, other aspects are also of consideration. One factor that is not reflected in an LCA is the work environment and demand for staff hours. Typically, a centrifuge is placed in a closed room. The centrifuging process causes heavy noise, odour and gas emissions inhaled by staff, and so protective clothing and relevant procedures are needed. Furthermore, accidents involving the active centrifuge can be serious. In comparison, the procedure for running an STRB system is very simple, in that the sludge is automatically loaded into the system through pipes, preventing staff from being in contact with the sludge. As the whole treatment process happens outside, gas emissions are not a concern in relation to health. Odour nuisance from STRB systems can be present, but only if the system is overloaded, while any noise related to the treatment process is minimal and of no concern. Hence, in terms of working environment and safety of the staff, STRB system technology performs better than mechanical treatment.

In the performed LCA, only the production of the polymer coagulant was included when calculating the environmental loadings provided by the centrifuging technology. The final sludge product produced by the conventional technology also contains traces of the added polymer coagulant; however, this is not reflected in the LCA results, as polymer traces are included in the impact categories. However, some European countries, such as Germany, are working on legislation against the presence of polymer traces in dewatered sludge intended for land application. If this legislation becomes a reality, STRB system technology will have a crucial advantage over mechanical treatment technologies.

Toxic impacts due to heavy metals were found to be the same for all three treatment scenarios. However, the effect of xenobiotics present in the final sludge products was not included in the impact categories addressed in this LCA. The contents of nonylphenol ethoxylates (NPE), di(2-ethylhexyl)phthalate (DEHP), linear alkylbenzene sulfonates (LAS) and polycyclic aromatic hydrocarbons (PAHs) in sludge products for land application are of concern, especially if threshold values in biosolids for land application for these compounds, defined by the Danish Ministry of Environment and Food (Miljøministeriet 2006), are not met. A study undertaken by the Danish Ministry of Environment and Food in 2000 (Miljø- og Fødevareministeriet 2000) found that degradation of the mentioned compounds is more efficient in sludge subjected to treatment in an STRB system compared to sludge that has been mechanically dewatered and stored.

Comparing the present LCA with other LCAs

In 2013, a study undertaken by the Ministry of Environment and Food of Denmark compared the environmental performances of 14 sludge treatment scenarios, among which were two scenarios comparable to S-CEN and S-STRB (Kirkeby *et al.* 2013). This study also found that eutrophication caused by N-containing compounds was higher for sludge dewatered on a centrifuge and subsequently stored compared to sludge treated in an STRB system, though other results, e.g. impacts on climate change, do not match the findings of our study. However, considerable parts of the inventory data used by Kirkeby *et al.* (2013), to model the environmental impacts caused by STRB systems, were not based on actual data from STRB systems but on data on emissions from crop land or compost windrows. Indeed, in a validation test of the data used in the study by Kirkeby *et al.* (2013), data available for STRB systems were granted the lowest score possible for reliability. Hence, the results presented in Kirkeby *et al.* (2013) are somewhat unreliable.

Uggetti *et al.* (2011), a Spanish study which compared the treatment of sludge in an STRB system with mechanical centrifuge treatment, found that STRB systems performed better compared to the mechanical treatment technology in impact categories defined as “Abiotic depletion”, “Acidification”, “Eutrophication” and “Global Warming”. In contradiction with our results, Uggetti *et al.* (2011) concluded that the impacts caused on “Climate Change” by emissions of CH_4 directly from sludge mineralisation subjected to treatment were negligible compared to the emissions of CO_2 caused by the consumption of electricity and fuel. However, this study did not include emissions of N_2O from STRB systems, while results of the present study shows that these emissions from mineralisation processes are highly relevant to include for both STRB systems and the mechanical treatment technology. Furthermore, Uggetti *et al.* (2011) did not include final disposal (land application), as the emissions related to this step were expected to be the same for all scenarios. The results of the present LCA found that this is not true; rather, emissions related to land application, especially those affecting marine eutrophication, are highly relevant when comparing the environmental performances of sludge treatment technologies. In addition, the LCA methodology and the data used in Uggetti *et al.* (2011) are somewhat non-transparent, thereby making it difficult to compare the outcome of that study with the outcome of the present study.

When evaluating the treatment efficiency and environmental performance of the STRB system technology it is important to have in mind that operational differences among different STRB systems could affect the outcomes considerably. The daily, operational procedures related to the STRB system technology are rather simple, however, if a STRB system for some reason becomes overloaded, the treatment efficiency and environmental performance is negatively affected. Therefore, when doing scientific research on STRB systems it is important to note the operational state of the reference system, such as the numbers of loading and resting days or the characteristics of the feed sludge. It is thereby possible to evaluate if the STRB system in consideration is well-operated and represents optimal performance, or if operational problems affect the results. Furthermore, the microbial activity responsible for the mineralisation processes happening during the treatment is highly affected by climate; hence, comparing the performance of STRB systems located in different climate zone should be done with caution. The dataset on the STRB system technology generated by the present project is representative for the STRB systems located in northern Europe; however, the performance of the technology could be different in other climate zones.

5 Conclusions and recommendations

The overall goal of the project was to perform an LCA comparing the environmental impacts of treatment of sludge in STRB systems with a mechanical technology, namely sludge centrifuging, and subsequent storage. Important secondary objectives were to provide reliable data supporting a sound environmental assessment.

In order to provide reliable data, knowledge gabs and three research areas defined, focusing on quantification of biological gas emissions from sludge treatment and storage, establishment of substance flows of different sludge treatment scenarios and determination of emissions from treated sludge when applied to the land. Data related to mechanical treatment of sludge were collected alongside with data for STRB systems.

For the LCA, three sludge treatment scenarios were defined: 1) treatment in an STRB system and finally land application (S-STBR), 2) treatment in an STRB system, followed by post-treatment on SPA and finally application (S-SPA) and 3) mechanically dewatering on centrifuge, followed by storage and finally land application (S-CEN). The LCI´s for the various scenarios were based on the data generated for the three research areas and operational data provided by a utility. An attributional LCA approach was chosen, and the loadings and savings for all impact categories were normalised to PE. The FU was defined as the environmental impacts caused by treatment of 1000 kg wet weight SAS.

Overall, the LCA revealed that the environmental impacts caused by the scenarios based on the STRB system technology were comparable to or lower than impacts caused by the scenario based on mechanical treatment and subsequent storage. For the impact category Climate Change the major part of the contributing loadings for all scenarios arose from biological activity in the sludge during treatment in STRB (S-STRB and S-SPA) or storage subsequent to mechanical dewatering (S-CEN). The research on gas emissions from biological processes happening in STRB systems revealed that seasonal variations in the emission rates are considerable, and therefore should be considered when calculating average, annual gas emission rates. Furthermore, the research revealed that the dynamics in the gas emission rates also follows the loading state of the bed.

The emission rates of CO_2 measured for STRB systems were much higher compared to those measured in stored, mechanically treated sludge, reflecting higher biologic activity in the sludge residing in the STRB system. However, as the emission rates of CH_4 and N_2O, and thereby percentage shares of carbon and nitrogen emitted as the strong greenhouse gasses CH_4 and N_2O, were larger for mechanical dewatered sludge, the net environmental loadings provided to the impact category Climate Change by this technology and the STRB technology ended up being equally sized ($9.0 \cdot 10^{-4}$ PE), despite of higher biological activity in the STRB systems.

For most other impact categories, the major environmental loadings caused by the three scenarios arose from applying the final sludge products to land. As all metals found in the sludge subjected to treatment in all scenarios eventually ended up on agricultural land, the toxic impacts caused by these metals were the same for all scenarios (the net-loadings for the impact categories Human Toxicity – Non-carcinogenic and Ecotoxicity being $2.0 \cdot 10^{-2}$ PE, and $5.0 \cdot 10^{-4}$ PE for Human Toxicity – carcinogenic). However, as the biological activity, and thereby the mineralisation of C and N, in sludge treated in STRB systems was higher compared to in mechanically treated and subsequently stored sludge, the final sludge product produced by mechanical treatment contained more C and N. Hence, the final sludge product produced by mechanical treatment had a larger eutrophication potential compared to the final sludge product produced by STRB systems, which was reflected in larger contributions to the impact categories related to eutrophication and acidification. This difference was especially pronounced for the impact category Marine Eutrophication, for which the net loading provided by mechanically treated sludge corresponded to $8.0 \cdot 10^{-3}$ PE, while it was $3.0 \cdot 10^{-3}$ PE for STRB systems.

Furthermore, for consumption of fossil and reserve abiotic resources the resulting environmental loadings were higher for the mechanical treatment technology, due mainly to a demand for polymer coagulant, but also to longer transport distances. As mechanically treated sludge often have a stronger odour compared to sludge treated in STRB systems, the latter is often claimed by the local land application sites, while mechanically treated sludge must be transported longer distances to land application sites willing to apply it. Hence, the STRB system technology required a lower input of fuel for transportation.

Environmental impacts caused by the scenarios based on treatment in an STRB system, excluding and including post-treatment at an SPA, respectively, were almost identical. However, not all aspects of the scenarios were fully expressed in the LCA: adding post-treatment in an SPA to STRB systems considerably shortens the time the emptied beds need to rest before they can be reintroduced into the loading scheme, which is an advantage in the longer run. Indeed, also when comparing STRB systems to mechanical treatment, aspects not directly reflected in the results of the LCA, such as work environment, should be considered.

The LCA provides a basis for decision-making in relation to which sludge treatment technology to employ in a given and specific situation. As eutrophication related to nutrient run-off from agricultural land is a heavily debated topic in Denmark, STRB systems have an advantage due to the efficient mineralisation of C and N compounds, especially because the effect on climate change is kept low. The magnitude of the environmental loadings related to the consumption of abiotic resources for all scenarios was small compared to the loadings caused by land application and biological greenhouse gas emissions.

6 Further research

- A more complete and reliable dataset on the STRB system technology for LCAs is now available; hence it would be relevant to compare the technology to more sludge treatment technologies commonly used, e.g. aerobic digesting.

- The dataset generated is representative for STRB systems located in the northern part of Europe. However, the technology is also widely employed in the southern part of Europe. Hence, a relevant future study would be to generate a similar dataset based on STRB system located in southern Europe, and also other parts of the world.

- The research on gaseous emissions from the biological processes in sludge subjected to treatment in STRB systems revealed that the seasonal variation in the emission rates was considerable. The gaseous emissions measured from mechanically treated and subsequently stored sludge did not cover all seasons. Hence, it would relevant to further investigate how seasonal variation affects gas emission rates from stored, mechanically treated sludge.

- The research on gaseous emissions from the biological processes in sludge subjected to treatment in STRB systems also revealed that the dynamics in gas emission rates also follows the loading state of the bed. In order to minimise the emissions of the potent greenhouse gasses CH_4 and N_2O it would be relevant to further investigate these dynamics.

- The substance flow analysis did not include the flow of xenobiotics. The contents of the xenobiotics NPE, DEHP, LAS and PAHs in sludge products for land application are of concern, as threshold values for the contents of these compounds in sludge for land application are defined by Danish and European legislation. Residues of polymer coagulant in sludge for land application are also of concern as some European countries are working on legislation on threshold values concerning such residues. Hence, it would be relevant to expand the substance flow analysis be including these substances.

- Economical assessments of the STRB system technology and other sludge treatment technologies have been carried out to some extent. However, more detailed economical assessments, based on the standards for LCA, would be a relevant topic for further investigation.

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8 Papers

I Larsen, J. D., Nielsen, S., Scheutz, C. 2017. Greenhouse gas emissions from the mineralisation process in a Sludge Treatment Reed Bed system: seasonal variation and environmental impact. *Ecological Engineering. In press.*

II Larsen, J. D., Nielsen, S., Scheutz, C. 2017. Gas composition of sludge residue profiles in a Sludge Treatment Reed Bed between loadings. *Water Science and Technology. Accepted for publication, May 2017.*

III Larsen, J. D., Nielsen, S., Scheutz, C. 2017. Assessment of Danish Sludge Treatment Reed Bed system and a stockpile area, using substance flow analysis. *Water Science and Technology. Accepted for publication, May 2017*

IV Gómez-Muñoz, B., Larsen, J. D., Bekiaris, G., Scheutz, C., Bruun, S., Nielsen, S., Jensen, L.S. Nitrogen mineralisation and greenhouse gas emission from the soil application of sludge from reed bed mineralisation systems. *Journal of Environmental Management. Under revision, May 2017.*

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VI Nielsen, S., Larsen, J. D. 2017. Operational technology, economic and environmental performance of Sludge Treatment Reed Bed systems–based on 28 years of experience. *Water Science and Technology*. DOI: 10.2166/wst.2016.295

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In this online version of the thesis, papers I-VI are not included but can be obtained from electronic article databases, e.g. www.orbit.dtu.dk or on request from:

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The Department of Environmental Engineering (DTU Environment) conducts science-based engineering research within six sections: Water Resources Engineering, Water Technology, Urban Water Systems, Residual Resource Engineering, Environmental Chemistry and Atmospheric Environment.

This project was conducted as a collaboration between DTU Environment and Orbicon A/S. Orbicon A/S is a Danish environmental consultancy providing consultancy, construction management and IT solutions on a wide range of topic categories related to green engineering.

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