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- Life cycle assessment comparing the treatment of surplus activated sludge in a sludge
- treatment reed bed system with mechanical treatment on centrifuge
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25 Abstract

In Denmark, the conventional method for treating sewage sludge is mechanical dewatering and 26 subsequent storage. However, sludge treatment reed bed systems, which are holistic sludge 27 treatment facilities combining the dewatering, mineralisation and storage of sludge, have been more 28 29 common during the last three decades. Treatment of sludge in a sludge treatment reed bed system can be combined with post-treatment (further dewatering and mineralisation) on a stockpile area. 30 This study aimed to compare the environmental performances of a mechanical sludge treatment 31 32 method with the sludge treatment reed bed system strategy, using the life cycle assessment approach and a life cycle inventory based on newly generated data obtained from Danish reference 33 facilities. The scenarios based on the different treatment methods were initiated by sludge entering 34 the sludge treatment reed bed system or the centrifuge and terminated by land application of the 35 final sludge product. The environmental impacts caused by the sludge treatment reed bed system 36 strategy was comparable to or lower than those caused by the mechanical sludge treatment method. 37 The impacts on climate change were the same for all the treatment scenarios; however, the 38 conversion of organic carbon and nitrogen into gas species was more efficient in the sludge 39 40 treatment reed bed system compared to mechanical treatment. Thus, mechanically treated sludge contained more nitrogen, causing higher nitrogen emissions (primarily nitrate run-off) when applied 41 on land. According to the results of the life cycle assessment, there were no considerable 42 43 environmental gains made by adding post-treatment on a stockpile area to the sludge treatment reed bed strategy. However, some practical aspects not included in a life cycle assessment, should also 44 be taken into consideration when evaluating the performances of sludge treatment scenarios. 45 46

47 Keywords:

48 Sewage sludge, land application, eutrophication, climate change, environmental impact, nitrogen

49 **1. Introduction**

Sludge Treatment Reed Bed (STRB) systems have been used for treating sludge in Denmark 50 since 1988 (Nielsen et al. 2014). The STRB system treatment method is also employed in other 51 European countries, e.g. France (Vincent et al. 2011), Italy (Peruzzi et al. 2013), Spain (Uggetti et 52 53 al. 2009) and United Kingdom (Nielsen & Cooper 2011). An STRB system is a holistic sludge treatment facility that combines the dewatering, mineralisation and storage of sludge. These systems 54 are often used for the treatment of sludge originating from domestic wastewater treatment, but they 55 are also used to treat other types of sludge, e.g. from waterworks (Nielsen & Cooper 2011) or aqua 56 cultural sludge (Summerfelt et al. 1999). Commonly, an STRB system is built as a part of a 57 wastewater treatment plant (WWTP) and receives the sludge produced at this unit. An STRB 58 59 system consists of a number of beds, often eight to 10 (Nielsen & Willoughby 2005) or more, to which sludge is applied over several years. While retained in the beds, the sludge is gradually 60 dewatered and mineralised. After 8 to 12 years of dewatering and mineralisation, the final sludge 61 product (the sludge residue left at the end of the entire treatment and storage processes) is excavated 62 and applied to agricultural land as fertiliser and soil improvement. 63

64 Since STRB systems were introduced in Denmark, it has been common practice to empty beds during late summer or early autumn and then transport the excavated sludge residue directly to 65 agricultural land after harvest. However, in recent years, a new procedure has been employed by 66 67 some STRB systems: The beds are emptied in spring and the excavated sludge residue immediately transferred to a stockpile area at the WWTP where it undergoes post-treatment (further dewatering 68 and mineralisation) until autumn. During post-treatment, the sludge residue undergoes further 69 70 mineralisation and dewatering due to increased evaporation. This approach has the advantage that the emptied beds can be put back into operation in summer, as the reeds will regrow during 71 spring/early summer. If the excavation happens in autumn, the emptied bed must still rest until next 72

coming spring/summer, as the reeds remain dormant during autumn and winter. Originally,

stockpile areas were of a simple design, namely an outdoor area on which the sludge residue was

piled. Recently, coverage of the area by a greenhouse roof and walls has been added to the design,

76 adding a solar drying effect to the post-treatment process.

Conventionally, sewage sludge is dewatered by mechanical devices, such as decanter 77 centrifuges and screw presses, and subsequently stored until it can be applied on agricultural land as 78 fertiliser (Jensen & Jepsen 2005). Only a few studies assessing the environmental impacts of sludge 79 80 treatment technologies include STRB systems (Uggetti et al. 2011; Kirkeby et al. 2013) has ben done, as data on STRB systems suited for life cycle assessment (LCA) are scarce. Furthermore, the 81 reliability of the results of these studies could be questioned: A considerable part of the inventory 82 83 data used by Kirkeby et al. (2013) to model the environmental impacts caused by the STRB system strategy, were not based on actual data from STRB systems but on emission data from crop land or 84 compost windrows. Hence, the results presented in Kirkeby et al. (2013) are somewhat unreliable. 85 The LCA method, data and assumptions used in Uggetti et al. (2011) are somewhat intransparent, 86 making it difficult to compare the outcome of that study with other studies. Only in recent years, 87 88 new life cycle inventory data on STRB systems, including substance flows in STRB systems (Larsen et al. 2017a), gas emissions from the mineralisation process occurring in STRB systems 89 (Larsen et al. 2017b) and fertiliser quality of sludge residue for land application (Gómez-Muñoz et 90 91 al. 2017), have been generated. Combined, these studies provide the first datasets on STRB systems made with the purpose of being suitable for LCA; these datasets were not available at the time the 92 mentioned studies by Uggetti et al. (2011) and Kirkeby et al. (2013) were conducted. 93

The aim of the present study was to evaluate the environmental performances of the STRB
system strategy using the newest obtained data and according to the international ISO standards for
LCA. The performance of the STRB systems strategy was compared to a conventional treatment

97 strategy based on centrifugal dewatering of sludge, subsequent storage and final application on agricultural land. To assure that the comparison of the treatment strategies was as up-to-date, new 98 99 process specific data for the conventional treatment strategy was generated for the purpose. Three sludge treatment scenarios, all of which reflected the management of surplus-activated sludge 100 (SAS) generated at a reference WWTP, were defined and covered treatment, storage, transportation 101 and application of the final sludge product on agricultural land, including the substitution of mineral 102 nitrogen (N), phosphorous (P) and potassium (K) fertiliser, and the treatment of reject water 103 104 generated during the sludge treatment process.

105

106 2. Materials and methods

107 *2.1. The life cycle assessment approach*

An LCA can be applied for comparing resource consumption and impacts on the environment 108 of products or services that provide the same function (ILCD 2010). The LCA in this study 109 complied with ISO 14040 and ISO 14044 standards. In this study, an attributional LCA modelling 110 approach was chosen, and the multi-functionality of processes was dealt with by employing system 111 112 expansion. LCA modelling was done with the mass flow-based LCA software EASETECH, developed by the Technical University of Denmark and as described in Clavreul et al. (2014). 113 The LCA included three sludge treatment scenarios based on specific case studies of the 114 115 sludge treatment methods employed at the main reference site, namely a WWTP in Helsinge (Denmark) (56°01'15N; 12°19,49E). This WWTP houses an STRB system, a stockpile area and a 116 mechanical sludge treatment device, namely a decanter centrifuge. Data for the life cycle inventory 117 118 (LCI) were collected at this site and supplemented by data from three other recent studies (Gómez-Muñoz et al. 2017; Larsen et al. 2017b, 2017a) carried out at Helsinge WWTP and another 119 comparable WWTP, namely Himmark WWTP, (Denmark) (55°2'44"N 9°45'55"E). 120

121	Helsinge WWTP receives domestic wastewater and treats it accordingly, corresponding to
122	25,000 person equivalents (PE) annually. The wastewater is treated by a mechanical-biological
123	wastewater treatment line. A more detailed description of the wastewater treatment line at Helsinge
124	WWTP is found in the Supplementary Information (SI) (section SI-1).
125	The STRB system at Helsinge WWTP was established in 1996 (Fig. SI-1a and 1b) and is a
126	representative reference for the present-day STRB system technology. Since 1996, it has been well
127	operated, delivering a final sludge residue of high quality. Table SI-1 provides an overview of the
128	operational data and system characteristics for Helsinge STRB.
129	The stockpile area at Helsinge WWTP was established in 2012 - 2013 (Fig. SI-1a and 1c) and
130	has a total area of 1,675 m^2 , with 800 m^2 covered by a greenhouse roof, which enhances
131	evaporation from the sludge residue subjected to treatment. Recently, greenhouse walls on two
132	sides have been added to the design.
133	
134	2.2. Scope definition
135	Three sludge treatment scenarios were analysed: 1) mechanical dewatering by a decanter
136	centrifuge, followed by six months of storage, 2) 12 years of treatment in an STRB system and 3)
137	12 years of treatment in an STRB system followed by four months of post-treatment at a stockpile
138	area. The scenarios were defined as:
139	
140	Scenario 1: Mechanical treatment (S-CEN)
141	Sludge is dewatered on a conventional decanter centrifuge and immediately transferred to a
142	container in which the sludge is stored for one week ("on-site storage"). Afterwards, the dewatered
143	sludge is transported 70 km by truck to an external sludge storage facility ("external storage").

144 Here, the dewatered sludge is laid out in layers 1 to 1.5 m in height on the floor in an enclosed

storage building. The dewatered sludge is not moved or treated during storage. The storage facility 145 continually receives sludge during the year until autumn, following which it is collected and 146 147 transferred to a land application site. This procedure implies that at the time of land application, some of the stored sludge has resided at the storage facility for almost one year, while some has 148 only been there for a few days; hence, the average storage time for this study was assumed to be six 149 months. Finally, the dewatered sludge is excavated from the storage facility, transported 200 km by 150 truck to a land application site and applied by tractor. 151 152 Scenario 2: Sludge treatment reed bed system (S-STRB) 153 Sludge is loaded into an STRB system and undergoes 12 years of treatment (more information on 154 the STRB system technology is provided in SI (section SI-2). The sludge residue (including reeds) 155 is excavated and immediately transported 10 km by truck to a land application site and applied on 156 land by tractor. 157 158 Scenario 3: Sludge treatment reed bed system and stockpile area (S-SPA) 159 160 Sludge is loaded into an STRB system and undergoes 12 years of treatment (the exact same procedure as in S-STRB). The sludge residue (including reeds) is excavated by an excavator and 161 transported 0.15 km by truck to a stockpile area. The sludge residue is piled and undergoes four 162 163 months of post-treatment, which is enhanced by solar drying. Finally, the final sludge product is excavated from the stockpile area, transported 10 km by truck to a land application site and applied 164 by tractor. 165 166 The system boundaries included all unit processes (Fig. 1) related to sludge treatment and

167 final land application, including the effect of fertiliser substitution, the treatment of reject water and168 the treatment of SAS produced from the reject water. The temporal scope for the emission inventory

and the impact assessment were both defined as 100 years, and the geographical boundary was
Denmark. The functional unit (FU) was defined as the treatment and disposal of 1000 kg wet
weight (WW) of SAS with characteristics corresponding to the SAS generated at Helsinge WWTP
(Table 1). We decided to base the FU on the WW of the SAS as a central purpose of the treatment
processes are dewatering, and thereby volume reduction of the sludge. If based on the dry weight of
the sludge, this aspect of the treatment processes would not be reflected in the results of the LCA.

175

176 2.3. Life Cycle Inventory (LCI)

177 2.3.1. Daily operation and transportation

Data and assumptions on daily operations, excavation and transportation included in the 178 179 various scenarios were based on the present-day situation and procedures at Helsinge WWTP. All scenarios included consumption of electricity due to pumping of sludge and reject water and the 180 consumption of fuel for excavation and transportation. Furthermore, the centrifuging process 181 included in S-CEN requires an additional input of electricity and that the sludge is pre-conditioned 182 by adding polymer coagulant; hence, emissions related to the production of polymer coagulant were 183 184 included in this scenario. Data on emissions related to the consumption of electricity and fuel in the different scenarios, and the production of polymer coagulants, were taken from the Ecoinvent v 3.3 185 database and the database included in the EASETECH software. More details on consumption by 186 the three scenarios are to be found in SI (section SI-3). 187

For S-CEN, it was assumed that the final sludge product was transported 200 km to the land application site, while in S-STRB and S-SPA this distance was only 10 km. Sludge residue treated in a well-operated STRB system commonly meets the threshold values for heavy metals and xenobiotics in biosolids for land application, as required by Danish legislation (Nielsen 2005; Miljøministeriet 2017). Furthermore, sludge residue is odourless. Therefore, sludge residue can

often be applied on agricultural land in the local area. On the other hand, sludge, which has been mechanically dewatered and subsequently stored, often has a strong odour. Even though the dewatered sludge meets the threshold values required by the legislation, the odour makes it difficult to find a land application site willing to take it (information provided by Grib Vand, the utility managing Helsinge WWTP). Therefore, longer transportation distances are often required, as there are fewer local land application sites available to receive the mechanical dewatered sludge.

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200 2.3.2. Gas emission rates and flow of substances included in S-CEN

Helsinge WWTP houses a centrifuge that is commonly used to treat SAS from other minor 201 WWTP's. To be able to model S-CEN, in which the SAS from Helsinge WWTP is dewatered on 202 203 the centrifuge, it was arranged that a batch of SAS was dewatered on the centrifuge, instead of being loaded into the STRB, and samples of SAS, dewatered sludge and reject water from the 204 centrifuging process were collected and characterised. These data were used to calculate the 205 amounts of substances allocated to reject water and dewatered sludge during the dewatering 206 process. Emission rates for carbon dioxide (CO_2) , methane (CH_4) and nitrous oxide (N_2O) 207 208 representing on-site and external storage were obtained from flux chamber measurements carried out at the on-site storage facility, a container, at Helsinge WWTP. The dewatered sludge was stored 209 in the container at a height of approximately 1.5 m for 100 days running from October to January. 210 211 Data on N₂ and ammonia (NH₃) emissions were estimated based on emission data recorded at Helsinge WWTP and data obtained from a study measuring gas emission rates from dewatered 212 sludge piled on an outdoor storage area at a Swedish WWTP (Samuelsson et al. 2018). The 213 214 Swedish WWTP has a wastewater treatment line that generates sludge comparable to the SAS generated by Helsinge WWTP and was therefore considered an appropriate reference. Gas emission 215

rates, losses of carbon (C) and nitrogen (N) and more information on data sources and calculationscan be found in SI (section SI-4).

The evaporation of water during on-site storage was assumed negligible, and during external 218 storage it was estimated by combining our calculated values for losing organic matter and data on 219 the total solids content found in dewatered sludge, before and after 200 days of storage, as 220 published in a publication by the Ministry of Environment and Food of Denmark (Miljø- og 221 Fødevareministeriet (2000). As this publication reports that no water leached from the sludge 222 residue during storage, and the amount of ash remained unchanged, it was assumed that no P, K or 223 metals had left the system. More information on data sampling procedures, calculations and the 224 shares of substances allocated to different streams in the treatment process can be found in SI 225 226 (section SI-4). An overview of the flow of substances in the S-CEN scenario based on an input of sludge corresponding to the FU (1000 kg WW SAS) is also provided in SI (section SI-6, Table SI-227 8). 228

229

230 2.3.3. Gas emission rates and flow of substances included in S-STRB and S-SPA

231 Losses of C and N by mineralisation, and the related gas emission rates during 12 years of treatment in the STRB system, were modelled using newly generated data presented by Larsen et 232 al. (2017b). In this study, gas emission rates (CO₂, CH₄ and N₂O) from the STRB system in 233 234 Helsinge, covering all four seasons of the year, were measured by employing static surface flux chambers. Ammonia is produced from NH₄⁺ and often constitutes a considerable part of the N loss 235 from sludge and slurries. However, in STRB systems, NH₄⁺ is quickly taken up by the reeds or 236 237 converted into NO₃⁻ through nitrification, thereby preventing the formation of NH₃. Therefore, it was assumed that the loss of N to NH₃ in STRB systems was negligible. Gas emission rates related 238 to the mineralisation process during four months of solar drying on stockpile area were modelled 239

based on measured emission rates of CO₂, CH₄ and N₂O, also through use of static surface flux

- chambers. C and N losses, gas emission rates and more information on data sources and 241 242 calculations are to be found in SI (section SI-5). The amounts of substances partitioned to reject water, final sludge residue, mineralisation and 243 evaporation during 12 years of treatment in an STRB and the four months of solar drying at a 244 stockpile area, were modelled based on the substance flow analysis presented in Larsen et al. 245 (2017a). The substance flow analysis presented in that study was based on another Danish STRB 246 system, namely Himmark STRB system, albeit it was assumed that the study was an appropriate 247 reference for our LCA, as both systems are run in accordance with the operational guidelines and 248 produce comparable sludge residues of high quality. More information on data sampling 249 250 procedures, calculations and the shares of substances allocated to different streams in the treatment process can be found in SI (section SI-5). An overview of the flow of substances in the S-STRB and 251 S-SPA scenarios based on an input of sludge corresponding to the FU (1000 kg WW SAS) is also 252 provided in SI (section SI-6, Table SI-8). 253
- 254

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255 2.3.4. Long-term emissions from land application and fertiliser substitution

Emissions related to land application of the final sludge products were modelled using 256 recently obtained emission data for N and C. The emission data representing soil application of SAS 257 258 treated in an STRB system (S-STRB) originate from a lab-scale soil incubation study presented in Gómez-Muñoz et al. (2017). The fate of C and N over a 100-year modelling period was obtained by 259 using the Daisy soil-plant-atmosphere system model (version 5.21). Gaseous emissions of NH₃ and 260 261 N₂O, leaching of NO₃⁻ to groundwater and surface water, N-uptake by crops and C sequestration were estimated. When sludge residues are applied on land, it reduces the need for mineral fertiliser. 262 The environmental savings related to avoiding the production and use of mineral fertilisers were 263

264	included in the LCA. It was assumed that ammonium nitrate substituted mineral N fertiliser, that
265	single superphosphate substituted P fertiliser and that potassium chloride substituted K fertiliser.
266	To obtain similar data representing the treatment of SAS in an STRB system combined with
267	post-treatment on a stockpile area (S-SPA) and dewatering by centrifuge (S-CEN), a similar
268	incubation study, following the exact same procedures as described in Gómez-Muñoz et al. (2017),
269	was conducted. More information on the modelling of emissions related to land application and the
270	savings from fertiliser substitution can be found in SI (section SI-7).
271	
272	2.3.5. Reject water treatment
273	The reject water generated from the centrifuge in scenario S-CEN or dewatering in an STRB
274	system in scenarios S-STRB and S-SPA was returned to the WWTP and treated along with

incoming wastewater, thus producing more SAS. All scenarios included one re-run of the reject
water, covering its pumping back to the WWTP, the wastewater treatment process (including
related emissions to the atmosphere and aquatic environments), the entire sludge treatment
processes for the different scenarios and final land application (including fertiliser substitution).

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280

2.4. Life cycle impact assessment (LCIA)

Mid-point impact potentials for 14 normalised impact categories were calculated: Depletion of Fossil Abiotic Resources; Depletion of Reserve-based Abiotic Resources; Climate Change; Marine Eutrophication; Freshwater Eutrophication; Terrestrial Eutrophication; Terrestrial Acidification; Stratospheric Ozone Depletion; Photo Oxidant Formation; Ionising Radiation; Particulate Matter Formation; Human Toxicology – Carcinogenic; Human Toxicology – Noncarcinogenic and Ecotoxicity. The choice of LCIA methods for the different impact categories was made according to recommendations provided by the International Reference Life Cycle Data

288 System (ILCD) (ILCD 2010; Hauschild et al. 2013). The normalisation reference was found in (Blok et al. 2013). LCIA methods and normalisation references are shown in SI (section SI-8). 289 290 The loadings and savings calculated for each impact category are presented in six subprocesses: 1) daily operation (electricity consumption for pumping sludge and reject water, polymer 291 coagulant consumption), 2) biological gas emissions during treatment and storage (gas emissions 292 related to on-site and external storage of centrifuged sludge or mineralisation processes in STRB 293 system and stockpile area), 3) transportation and excavation (fuel consumption for trucks, 294 295 excavators and tractors), 4) land application (gaseous emissions and leaching of substances related to land application of the final sludge product), 5) fertiliser substitution (the effect of substituting 296 the production and use of mineral fertiliser) and 6) reject water treatment (RWT) (pumping of reject 297 298 water back to the WWTP, electricity consumption related to treatment, gaseous emissions and leaching related to treatment, the re-running of the produced SAS through the entire sludge 299 treatment process, including land application and fertiliser substitution). 300

301

302 2.5. Uncertainty analysis

The robustness of the results was analysed on two levels. First, a contribution analysis was 303 performed to identify substances influencing more than 90% of the overall environmental impact; 304 the results are shown in Table 2. Second, a sensitivity analysis (SA) was conducted by increasing 305 306 and decreasing mineralisation rates and transportation distances for all scenarios. SA-1 tested how increasing or decreasing the C and N mineralisation rates in all scenarios by 10% of its original 307 value affected the outcome of the LCA. SA-2 tested how changing the transport distances affected 308 309 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 did not interfere. 310

312 **3. Results and Discussion**

313 3.1. Impact categories

The results of 11 of the 14 impact categories are shown in Fig. 2. The results of the three impact categories not included in Fig. 2 (Stratospheric Ozone Depletion, Photochemical Oxidant Formation and Ionising Radiation) are shown in SI (section S1-9). The impacts of these categories were low compared to the impact categories shown in Fig. 2, and therefore they will not be discussed further.

319

320 *3.1.1. Climate change, eutrophication and acidification*

For the impact category Climate Change, the scenarios provided almost equal net loadings 321 322 into the environment. The loadings adding to this impact category is due mainly to CH₄ and N₂O emissions from treatment in the STRB system (S-STRB and S-SPA), post-treatment at the stockpile 323 area(S-SPA) and storage of dewatered sludge (S-CEN) (Fig. 2b and Table 2). However, in all 324 scenarios emissions of CH₄ and N₂O from the final sludge products after being applied on land also 325 added considerable to Climate Change; indeed, for S-CEN almost 50 % of the loadings adding to 326 327 this category was caused by emissions of CH₄ and N₂O related to land application. Methane and N₂O are strong greenhouse gasses having global warming potentials (GWPs) corresponding to 28 328 and 265 CO₂ equivalents, respectively, and therefore important to consider in relation to Climate 329 Change (IPCC 2014). 330

In all scenarios the treatment processes and the land application processes also emitted CO_2 and N_2 . However, 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), these emissions are climateneutral. During the 12-year treatment process in the STRB system in S-STRB and S-SPA, the main share of the mineralised C and N was emitted as CO_2 (93%) and N_2 (94%), while the remaining

336 shares of mineralised C (7%) and N (6%) were emitted as CH₄ and N₂O, respectively. On the other hand, during the six months of storage of centrifuged sludge at the on-site and external storage 337 facilities in S-CEN, only 48% of the C mineralised and 74% of the N mineralised was emitted as 338 climate-neutral CO₂ and N₂, while the remaining shares of C (52%) and N (26%) were emitted as 339 CH₄ and N₂O. The greater production and emission of CO₂ and N₂ from the STRB system in S-340 STRB and S-CEN compared to the sludge storage facilities is assigned to the efficient aeration of 341 the sludge residue in the STRB system: Air leak to the sludge residue through rhizomes (hollow 342 out-growths produced by the reeds), movements of stems create cracks in the sludge residue surface 343 through which air enters and the joint reject water pipe and ventilation system embedded in the 344 filter layer provides air to the lower parts of the sludge residue (Nielsen 2003). Aeration enhance 345 346 aerobic microbial activity, leading to the production of CO₂ and N₂. At the sludge storage facilities centrifuged sludge was not moved or turned during the storage period. An earlier study found that 347 anaerobic conditions are prone to develop in dewatered sludge stored in a storage facility without 348 being turned (Nielsen, 2005), leading to production of CH_4 and N_2O . 349

Aerobic mineralisation is more effective compared to anaerobic mineralisation in terms of 350 351 the amount of C and N converted into gas species. Hence, the amounts of C and N mineralised during treatment in the STRB system were almost twice the amounts mineralised during storage of 352 the centrifuged sludge (Table SI-8), while the emissions to air impacting Climate Change provided 353 354 by biological gas emissions were almost the same for all three scenarios (Fig. 2). Hence, C and N is more efficiently removed from the sludge subjected to treatment in S-STRB and S-SPA compared 355 to the sludge treated in S-CEN, despite of the impacts on Climate Change are equal for all three 356 357 scenarios.

The amount of C and N found in the final sludge product affected the impacts from greenhouse gas emissions related to land application. The slower mineralisation rate during the

360 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 on land. Indeed, the greenhouse gas emissions from land 361 application of sludge residue were higher for S-CEN compared to S-STRB and S-SPA (Fig. 2). The 362 share of N emitted as N₂O after soil application was approximately 3% for all three sludge products 363 (SI Table SI-9); however, N content in the centrifuged, final sludge product was greater compared 364 to the sludge residue from the STRB system, leading to a larger contribution to Climate Change 365 from S-CEN compared to S-STRB and S-SPA. For all scenarios, small environmental savings in 366 Climate Change impact category were obtained by substituting mineral fertiliser. 367 For Marine Eutrophication, all scenarios showed a net loading, mainly caused by NO₃⁻ 368 leaching and run-off from land application (Fig. 2b and Table 2). The impact caused by S-CEN was 369 370 more than twice the impacts caused by S-STRB and S-SPA. This higher loading in S-CEN was due to the larger N content in the centrifuged sludge, and higher emission factors for NO₃⁻ leaching and 371 run-off. 372

For Terrestrial Eutrophication, all scenarios showed a net loading, caused primarily by NH₃ emissions from the land application process (all scenarios) and by NO_x and N from the combustion of fossil fuels (all scenarios, but especially S-CEN) (Fig. 2b and Table 2). Ammonia emissions were highest in S-CEN, as the final sludge produced in this scenario contained more N and had a larger NH₃ emission rate for land application than the final sludge in the other scenarios (S-STRB and S-SPA).

The impact category Terrestrial Acidification was affected primarily by NH_3 emissions from land application and by SO_x and NO_x from the combustion of fossil fuels (Fig. 2b and Table 2). For S-CEN and S-STRB, the overall impacts were small net loadings. For S-SPA, the overall impact was a net saving, as the savings caused by fertiliser substitution exceeded the loadings caused by the other sub-categories.

384	For Freshwater Eutrophication, net loadings were seen for all scenarios (Fig. 2b). For S-STRB
385	and S-SPA, these loadings were caused mainly by phosphate (PO_4^{3-}) leaching from the land
386	application of sludge residues, while for S-CEN the impact potentials caused by PO_4^{3-} leaching
387	from land application and reject water treatment were equal in size (Fig. 2b and Table 2). The
388	concentration of P in the reject water produced by the centrifuge was 10 times the concentration
389	identified in the reject water produced by the STRB. The P leaching factor from the WWTP was
390	relatively high compared to the leaching factor from land application. Therefore, the impact to
391	Freshwater Eutrophication in S-CEN was higher than in S-STRB and S-SPA.
392	
393	
394	
395	3.1.2. Ecotoxicity and human toxicity (non-carcinogenic)
396	Among all the impact categories, the most affected were Ecotoxicity and Human Toxicity –
397	Non-carcinogenic (Fig. 2a). However, metals diverted to the reject water in S-CEN are eventually
398	also land applied, leading to that the contribution to Human Toxicity – Non-carcinogenic and
399	Ecotoxicity were equal for all scenarios. For both impact categories, all scenarios provided a net
400	loading, caused primarily by the presence of zinc and copper in the final sludge product when
401	applied on land (Table 2). As these impact categories are affected by the same substances, the
402	overall results are the same, except for the magnitude of the values. For both categories, the net
403	loadings were the same for all scenarios. In S-STRB and S-SPA, the loading caused by land
404	application was slightly higher compared to S-CEN, due to the larger amounts of metals transferred
405	to the final sludge product produced in S-STRB and S-SPA (Section SI-6, Table SI-8 in SI).
406	

408 *3.1.3. Human toxicity (carcinogenic), resource depletion and particulate matter*

For all scenarios, Human Toxicity – Carcinogenic was affected primarily by the presence of
nickel and lead in the final sludge product when applied on land (Fig. 2c and Table 2). Small
savings were provided by fertiliser substitution. As for Human Toxicity – Non-carcinogenic and
Ecotoxicity, the overall impacts were similar for all scenarios.

The impact of Depletion of Fossil Abiotic Resources (Fig. 2c) was higher for S-CEN than for 413 the other two scenarios (S-STRB and S-SPA), due to larger fossil fuel demand related to daily 414 operation, transportation and excavation in this scenario. The main impacts were caused by the 415 consumption of hard coal and crude oil, while the main impacts in S-STRB and S-SPA arose solely 416 from the consumption of hard coal (Table 2). For S-STRB and S-SPA, the overall results were 417 418 small net savings, as savings from the substitution of mineral fertiliser exceeded loadings. For S-CEN, daily operations included the production and consumption of polymer coagulant required for 419 pre-conditioning the sludge prior to centrifuging. The production of this polymer coagulant caused 420 the consumption of crude oil and the higher environmental loading. Furthermore, the transport 421 distances, earlier addressed in section 2.3.1, included in S-CEN were 70 km from the WWTP to the 422 423 external storage facility, followed by 200 km to the land application site, compared to 0.150 km from the STRB system to the stockpile area in S-SPA, and 10 km to the land application sites in S-424 STRB and S-SPA, resulting in a considerably greater demand for fuel in S-CEN. 425

For the impact category Depletion of Reserve-based Abiotic Resources, all scenarios showed net savings, as the resource consumption avoided from the substitution of mineral fertiliser exceeded the resources needed for sludge management. In S-STRB and S-SPA, positive loadings were negligible compared to savings. For S-CEN, loading was caused mainly by the consumption of lead in relation to the consumption of crude oil. For S-CEN, savings as a result of fertiliser substitution were slightly greater compared to S-STRB and S-SPA, as more mineral fertiliser was

432	assumed to be replaced in this scenario. However, due to the larger loading in S-CEN, this scenario		
433	provided a lower net saving compared to the other scenarios.		
434	For Particulate Matter, all scenarios showed net savings, due to the substitution of mineral		
435	fertiliser. Positive contributions arose mainly from emissions of NH_3 and sulphur dioxide (SO ₂)		
436	related to the combustion of fuel, and NH ₃ emissions from land application.		
437			
438	3.2. Sensitivity analysis		
439	3.2.1. Mineralisation rates		
440	In SA-1, the mineralisation rates for C and N mineralised were increased and decreased by		
441	10% of their original values in all three treatment scenarios (Fig. 3). Changes in the mineralisation		
442	rates during the treatment of sludge in the STRB system, in the stockpile area or while storing		
443	mechanical dewatered sludge at the external storage facilities affected the impact category Climate		
444	Change, as CH_4 and N_2O emissions were affected (Fig. 3). Furthermore, changing the		
445	mineralisation rates affected Marine Eutrophication, as the amount of N found in the final sludge		
446	product from the various scenarios depended on the amount of N mineralised earlier in the		
447	treatment process. The effects of SA-1 on the remaining impact categories can be found in SI		
448	(section SI-10).		
449	For Climate Change, S-CEN was more affected by changes in the mineralisation rates than		
450	the other two scenarios (S-STBR and S-SPA) (Fig. 3). The reason for this was that a larger share of		
451	the C and N mineralised in S-CEN was emitted as CH ₄ and N ₂ O than in the other two scenarios		

452 (Table SI-4 in section SI-4 and Table SI-6 in section SI-5 in SI). When the mineralisation rates for

453 C and N were decreased by 10% of their original values for all scenarios, S-CEN showed a lower

454 Climate Change impact than the other scenarios, while it was higher in the default scenario and

455 when mineralisation rates were increased by 10% (Fig. 3).

456	Changing the mineralisation rates for C and N had only a small effect on the impact of Marine
457	Eutrophication in the S-CEN (Fig. 3). Due to a much higher mineralisation rate in the STRB system
458	than while storing centrifuged sludge, S-STRB and S-SPA showed changes in Marine
459	Eutrophication. With a 10 % higher mineralisation rate, less N remained in the sludge product,
460	leading to a lower impact on Marine Eutrophication, while with a 10 % lower mineralisation rate,
461	more N remained in the sludge product, leading to a higher Marine Eutrophication impact.
462	However, regardless of the mineralisation rate applied, the impact on Marine Eutrophication impact
463	caused by S-CEN was always more than twice as high compared to S-STRB and S-SPA.
464	The results of SA-1 reflect a trade-off between the impact on Climate Change and on Marine
465	Eutrophication for the mineralisation rates of C and N during treatment or storage. Higher
466	mineralisation rates led to a higher Climate Change impact for S-CEN, but a lower Marine
467	Eutrophication impact for S-STRB and S-SPA, while lower mineralisation rates had the opposite
468	effect.

469

470 *3.2.2. Transport distances*

In SA-2, transport distances in the various scenarios were changed. The total transport 471 distances included in the various scenarios were 270 km for S-CEN, 10 km for S-STRB and 10.15 472 km for S-SPA. First the transport distances were halved for all the scenarios. Second, the transport 473 distance included in S-CEN was reduced to 10.15 km, to match the transportation distance in the 474 475 other scenarios. The impact category mainly affected by these changes was Depletion of Fossil Abiotic Resources. The effects of SA-2 on the impact category Depletion of Fossil Abiotic 476 477 Resources are shown in Fig. 3, while the effects on the remaining impact categories can be found in section SI-10 in SI. 478

For S-STRB and S-SPA, halving the transport distances did not affect the net impact on Depletion of Fossil Abiotic Resources (Fig. 3), while the net impact for S-CEN was reduced by almost 50%. However, despite this reduction, the impact potential of S-CEN remained higher than for the other scenarios. Reducing the transportation for S-CEN so it equalled the transportation distance in STRB and S-SPA drastically decreased the impact on Depletion of Fossil Abiotic Resources for S-CEN, though it remained greater than in the other two scenarios, due to a high contribution from producing polymer coagulant.

486

487 *3.3. Discussion*

The results of the LCA revealed that in terms of eutrophication of marine environments, the 488 489 treatment scenarios based on the STRB system strategy (S-STRB and S-SPA) caused lower impacts compared to the conventional strategy using mechanical dewatering on centrifuge (S-CEN). This 490 difference between the treatment strategies are mainly due to that treatment in STRB systems 491 provides a fuller mineralisation of C- and N-containing compounds, without causing a higher 492 emission of greenhouse gasses. This means that none of the strategies are more favourable when 493 494 considering impacts on climate change; however, the STRB system strategy is favourable in terms of avoiding eutrophication of marine environments in costal zones. Eutrophication of costal zones 495 due to nutrient run-off from agricultural land has during the last decades been a major problem in 496 497 Denmark, meaning that this difference between the strategies is highly relevant in Denmark and other countries with similar environmental problems. 498

The LCA study presented in Kirkeby et al. 2013 also found that eutrophication caused by Ncontaining compounds was higher for sludge treated by a conventional strategy based on
mechanical dewatering and subsequent storage compared to sludge treated in an STRB system.
However, due to a lack of data for the STRB system strategy at the time Kirkeby *et al.* (2013)

conducted their study it difficult to make valid conclusions based a comparing those results to thoseof our study.

Uggetti et al. (2011), a Spanish study comparing the treatment of sludge in a STRB system 505 with dewatering on centrifuge, did not include emissions of N₂O from the STRB systems, while the 506 results of our study show that emissions of N₂O from mineralisation processes are highly relevant to 507 include for both the STRB system method and the mechanical treatment method. Furthermore, 508 Uggetti et al. (2011) did not include final disposal (land application), as the emissions related to this 509 step were expected to be the same for all scenarios. The results of our LCA suggests that this is not 510 true but that emissions related to land application are highly relevant when comparing the 511 environmental performances of sludge treatment methods. 512 513 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 were not included 514 in the impact categories addressed in this LCA. The contents of nonylphenol ethoxylates (NPE), 515 di(2-ethylhexyl)phthalate (DEHP), linear alkylbenzene sulfonates (LAS) and polycyclic aromatic 516 hydrocarbons (PAHs) in sludge products for land application are of concern if the threshold values 517 518 for these compounds, defined by the Danish Ministry of Environment and Food (Miljøministeriet 2006), are not met. Hence, the flow of xenobiotics in the treatment scenarios would be a relevant 519

520 topic for future studies.

521 Overall, the environmental impacts of S-STRB and S-SPA are almost the same. However, 522 adding post-treatment on to stockpile area to the STRB system strategy has some practical 523 advantages that are not expressed in the results of the LCA. The presence of a stockpile area makes 524 it possible to empty STRB system beds in spring, thereby allowing the reeds in the excavated bed to 525 regrow within a few months, compared to almost one year if excavation happens in autumn. Faster 526 regrowth of the reeds implies that the bed can be reintroduced faster into the loading cycle with a

full loading programme, which enhances the treatment capacity of the STRB system. The stockpile
area also provides more flexibility in terms of time for excavating and collecting the final sludge
product by the recipient.

530

531 **4. Conclusions**

The environmental impacts caused by the sludge treatment scenarios based on the STBR 532 system strategy performed comparable or better compared to the scenario representing a 533 534 conventional sludge treatment strategy including mechanical dewatering on a centrifuge and subsequent storage. Carbon and nitrogen was more efficiently removed by the treatment processes 535 included in S-STRB and S-SPA, resulting in a lower content of C and N in the final sludge product 536 537 compared to S-CEN, despite of the impacts on Climate Change caused by gas emissions from the treatment process were equal for all scenarios. The lower content of C and N in the final sludge 538 product produced by S-STRB and S-SPA resulted in considerable lower impacts on Marine 539 Eutrophication compared to S-CEN. A sensitivity analysis revealed that the performances of S-540 STRB and S-SPA were more robust to changes in the amounts of C and N mineralised during the 541 542 treatment process and changes in transport distances compared to S-CEN. In terms of human toxicity and ecotoxicity, the impacts for all three treatment scenarios were comparable. According 543 to the results of the LCA, there were no considerable differences in the performances of S-STRB 544 545 and S-SPA. However, adding a stockpile area to the STRB system strategy had some practical advantages, which should be considered. 546

547

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- 554

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Table 1. Quality of the surplus-activated sludge produced at the Helsinge wastewater treatment

 plant (WWTP).

Wastewater treatment at Helsinge WWTP				
Sludge type		Surplus-activated Sludge	(SAS)	
Sludge age (aerobic days)		20-25		
Phosphorous removal		PIX		
Characterisation of surplus-activated sludge (SAS)				
Parameter		Parameter		
Total solid (TS) (% of WW)	0.6790	Cr (% of DW)	0.0023	
Volatile solid (VS) (% of DW)	61.483	Mn (% of DW)	0.0747	
Total nitrogen (TN) (% of DW)	3.9700	Fe (% of DW)	6.3970	
Total carbon (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)	0.00000001	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	

WW: wet weight, DW: dry weight

Table 2. Compounds responsible for > 90% of the total impact in 11 impact categories for the three scenarios. The compounds vary among the following six life cycle stages: daily operation, biological gas emissions, transportation/excavation, land application, fertiliser substitution and reject water treatment (RWT).

Impact Category	S-CEN	S-STRB	S-SPA
Climate change	CH_4 , N_2O	CH_4 , N_2O	CH_4 , N_2O
Freshwater eutrophication	PO ₄ ³⁻ , P	PO ₄ ³⁻ , P	PO ₄ ³⁻ , P
Marine eutrophication	NO_3^-	NO_3^-	NO ₃ ⁻
Terrestrial acidification	NH ₃ , SO _x , NO _x	NH ₃ , SO _x , NO _x	NH ₃ , SO _x , NO _x
Terrestrial eutrophication	NH ₃ , NO _x	NH ₃ , NO _x	NH ₃ , NO _x
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- based abiotic resources	In, Cd	In, Cd	In, Cd
Particulate matter	NH ₃ , SO ₂	NH_3 , SO_2	NH ₃ , SO ₂
Photochemical oxidant	NO _x	NO _x	NO _x
formation	NMVOC	SO_2	SO_2
Stratospheric ozone depletion	CFC-11, CFC-13, HCFC-12	CFC-11	CFC-11

CER

[Type text]



Fig. 1. Unit processes for three sludge treatment scenarios: Scenario S-CEN (dewatering on a centrifuge, one week of on-site storage and 6 months' external storage until land application), Scenario S-STRB (12 years of treatment in an STRB system, excavation in autumn and immediate application on land) and Scenario S-SPA (12 years of treatment in an STRB system, excavation in spring, four months' solar drying at an SPA and, finally, application on land during the following autumn).







Fig. 2. Life cycle impact assessment of 11 impact categories for three treatment scenarios: 1) dewatering on a centrifuge (S-CEN), 2) 12 years of treatment in STRB (S-STRB) and 3) 12 years of treatment in STRB, followed by four months of post-treatment in a stockpile area covered by a greenhouse roof (S-SPA).



Fig. 3. Results of the sensitivity analysis (SA) testing the robustness of the results in relation to mineralisation rate (SA-1) and transport distances (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" from the different scenarios, if the amounts of mineralised C and N are decreased or increased 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.

1 Highlights:

- 2 A life cycle assessment comparing sludge treatment scenarios was performed
- The assessment focused on environmental impacts related to 14 impact categories
- One scenario was based on mechanical dewatering, two on treatment in reed beds
- Newly generated process specific inventory data was used to model the scenarios
- Overall, the treatment in reed beds performed better than mechanical dewatering

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