

Sludge quality after 10–20 years of treatment in reed bed systems

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Received: 25 August 2014 / Accepted: 3 November 2014 / Published online: 26 November 2014
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Abstract The effect on the environment of the operation of sludge treatment in reed beds (STRB) system is seen as quite limited compared to traditional sludge treatment systems such as mechanical dewatering, drying and incineration with their accompanying use of chemicals and energy consumption. There are several STRB systems in Denmark receiving sludge from urban wastewater treatment plants. Stabilization and mineralization of the sludge in the STRB systems occur during a period between 10 and 20 years, where after the basins are emptied and the sludge residue typically is spread on agricultural land. In the present study, the sludge residue quality after treatment periods of 10–20 years from four Danish STRBs is presented. After reduction, dewatering and mineralization of the feed sludge (dry solid content of 0.5–3 %) in the STRB systems, the sludge residue achieved up to 26 % dry solid, depending on the sludge quality and dimensioning of the STRB system. The concentration of heavy metals and hazardous organic compounds in the sludge residue that are listed in the Danish and EU legislation for farmland application of sludge was below the limit values. The nitrogen and phosphorus concentrations as an average in the sludge residue were 28 and 36 g/kg dry solid (DS), respectively. In addition, mineralization on average across the four STRB systems removed up to 27 % of the organic solids in the sludge. The investigation showed that the sludge residue qualities of the four STRBs after a full treatment period all complied with the Danish and European Union legal limits for agricultural land disposal.

Keywords European Union legal limits · Sludge reed bed system · Sludge residue · Fertilizer · Hazard organic compounds · Heavy metals

Introduction

Sewage sludge is a by-product of wastewater treatment in wastewater treatment plants. In the last decades, there has been growing focus on the treatment and handling of the enormous amounts of sludge produced and on the negative effects that may be related with the disposal of sludge in the environment. In Denmark, municipal wastewater treatment plants (WWTPs) produce over 1.3 million tons of sludge every year. The total dry solid (DS) content in the sludge produced from municipal wastewater treatment plants has in the period from 1995 to 2009 ranged from 132,000 to 162,000 t DS (latest available data, Danish Environmental Protection Agency 2012, Kirkeby et al. 2013). In Denmark, sludge from WWTPs is primarily dewatered by mechanical treatment or in sludge treatment reed bed (STRB) systems.

The primary final disposal for the dewatered sludge in Denmark is on agricultural land. For instance, in 2009, 144,000 t DS was produced, and of this, more than half was spread directly on agricultural land (75,000 t DS; 52 %) or applied to STRB systems for later application on agricultural land (17,000 t DS; 12 %). The remainder of the 144,000 t DS was either incinerated (34,000 t DS; 24 %) or exported/put in landfills (16,000 t; 12 %) (Danish Environmental Protection Agency 2012, Kirkeby et al. 2013). Before any land application of mechanically treated sludge, the sludge has to be stored for hygienization reasons for a minimum period 6 months. In STRB, the hygienization process of the sludge occurs during the treatment period in the basins.

There exist several technologies for sludge treatment and dewatering on the market ranging from traditional mechanical

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treatment by decanter and screw-press technology to the environmentally friendly, but more area demanding, STRB systems. STRBs are also reported to be very cost-effective in daily operation compared to that of traditional mechanical solutions (Uggetti et al. 2011, 2012). Opposite most mechanical treatment, the dewatering of sludge in STRB occurs without the use of chemicals and with a minimum of energy consumption.

The treatment of wastewater sludge in STRB systems are a widespread and common sludge treatment practice in northern Europe and especially Denmark, where the technology has been used for more than 26 years (since 1988, Nielsen et al. 2014). The treatment period in STRB is approximately 10–12 years, but up to 20 years has been obtained (Nielsen et al. 2014).

During treatment in the STRB system, sludge with an initial dry solid content of 0.5–3 % is dewatered by drainage, evapotranspiration and mineralization processes. After final treatment, the sludge residue will typically end up having a dry solid content from 20 % and up till 35 % depending on the sludge quality treated in the systems (Nielsen 2011). Due to these mechanisms, the fresh sludge volume is reduced up to 98 %. The mineralization processes during the treatment period also influence the sludge quality. Typically, a gradient in the sludge residue profile is observed. The final sludge product in the oldest layers has lower concentrations of organic solid (per kg DS), while the relative content of more recalcitrant compounds and heavy metals (per kg DS) may have increased during the treatment period (Matamoros et al. 2012).

The content of hazardous organic compounds in sludge foreign to the environment usually can be reduced to such a degree that the sludge conforms to the limits and norms for deposition on agricultural land. After the treatment of sludge, recycling options are good, particularly in agriculture. The sludge quality is cleaner and more adaptable in the natural cycle than mechanically dewatered sludge (Nielsen 2005).

Danish legislation for sludge application

In order to apply sludge on farmlands in Denmark, the following criteria have to be fulfilled (Danish legal legislation anno 2014):

- 1) The sludge quality has to comply with the Danish legal limits for sludge application listed in the national guidelines (Table 1).
- 2) Suitable farmland has to be found (easier in autumn than in the spring, due to more available farmland after harvest).

If the above conditions are met, the permissible application rate of sludge is determined by the sludge's P and N content

(Danish legislation for sludge, BEK No. 1650 of 13 December 2006). The maximum allowable amount of phosphorous is 90 kg total phosphorus (TP)/ha every third year and for nitrogen 170 kg total nitrogen (TN)/ha/year. Moreover, the total nitrogen (N) and phosphorus (P) contents applied with the sludge have to be accounted for in the farmers' manure plan.

During the 1990s and onwards, stricter legislation has been brought into effect by the Danish Environmental Protection Agency (DEPA) to regulate the content of nutrients, heavy metals and hazardous organic compounds in sludge being spread on agricultural land (Nielsen 2005). For instance, feed sludge since February 2000 has to be analyzed for heavy metal content (ranging from 1 to 18 times/year depending on sludge quality and tons of DS production) and comply with the Danish standards listed in the Danish legislation for sludge (BEK No. 1650 of 13 December 2006) before being loaded to agriculture or STRB systems (Table 1). As an example, sludge with a chrome concentration corresponding to 60 % of the legal limit values and a yearly production of 500 t DS will require four analyses/year. Thus, it is not obligatory to analyze for heavy metals in the sludge residue when emptying the basins, as this has already been done during sludge loading of the STRB.

For certain heavy metals (cadmium (Cd), nickel (Ni), lead (Pb), mercury (Hg)), legal limit values for the application to farmland are related to both dry solid content (mg per kg DS) and to the total phosphorous (mg per kg TP) content in the sludge (Table 1). However, the sludge quality only has to comply with *one* of the limit values—i.e. if the Cd content in the sludge is exceeded when related to dry solid content, but is below the limit value when related to the TP content, sludge is allowed to be applied to agricultural land. The reasoning behind this is that commercial phosphor (NPK)-fertilizer also contains heavy metals and thus, when spread onto farmland, apply heavy metals to the environment likewise sludge. In the Danish legislation, the upper limits for the selected heavy metals in sludge correspond to the approximate concentration of heavy metals that would be spread, if the sludge-P was to be substituted by a similar amount of fertilizer P.

In EU, several member states have enacted and implemented stricter limit values for heavy metals and set requirements for other contaminants in sludge bound for agricultural land. Compared to EU legislation for sludge, Danish legislation has stricter requirements with several times lower cut-off limit values for heavy metals such as cadmium (Cd), mercury (Hg) and chromium (Cr) (Table 1). Contrary to current EU legislation, Danish legislation also sets up limits for hazardous organic compounds (linear alkyl benzene sulfonates (LASs), nonylphenol/nonylphenol ethoxylates (NPEs), di-2-ethylhexyl-phthalates (DEHPs) and polyaromatic hydrocarbons (PAHs)). Before application to agricultural land in Denmark, the sludge residue always has to be analyzed for

Table 1 The Danish and European Union legal limits for heavy metals and hazard organic compounds in sludge residue for agricultural use

Limit values	Denmark		EU	
	BEK No. 1650 of 13/12/2006		86/278/EEC EU Directive	ENV. E 3 (2000) Working document on sludge., 3rd draft
Heavy metals	mg/kg DS	mg/kg TP	mg/kg DS	mg/kg DS
Cadmium (Cd)	0.8	100	20–40	10
Copper (Cu)	1000	–	1000–1750	1000
Nickel (Ni)	30	2500	300–400	300
Lead (Pb)	120	10,000	750–1500	750
Zinc (Zn)	4000	–	2500–4000	2500
Mercury (Hg)	0.8	200	16–25	10
Chromium (Cr)	100	–	–	1000
Org. contaminants	mg/kg DS	mg/kg TP	mg/kg DS	mg/kg DS
LAS	1300	–	–	2600
PAH	3	–	–	6
NPE	10	–	–	50
DEHP	50	–	–	100

hazardous organic compounds. Currently, the EU Environment is working on a proposal including limits for hazardous organic compounds for a new EU directive with stricter limits (ENV. E3 (2000); working document on sludge, 3rd draft), but it has not been adopted yet. Sludge treatment reed bed system is mentioned as a sludge treatment method in the proposal.

Several STRB systems in Denmark have been in operation for more than 15–20 years and have been through one emptying period and some have been through a second emptying period. However, only limited information has been published about the sludge residue quality after a full treatment period in the STRB basins (e.g. Nielsen et al. 2014).

The present study presents data on the sludge residue quality after 10–20 years of treatment from four STRB systems situated in Denmark (Helsingø, Nakskov, Kallerup and Kolding) with the objective to describe the sludge residue quality after a full treatment period in relation to the Danish and EU legislation for sludge. Does the sludge residue comply with the legislation standards and cut-off limits for sludge application on agricultural land?

Methods

Treatment principle of sludge treatment reed bed systems

The overall sludge reduction takes place in the reed-planted basins in two ways; partly due to dewatering (draining, evapotranspiration) and partly due to mineralization of the organic solids in the sludge. Sludge from wastewater treatment plants is pumped onto the sludge residue surface in the basins. The dewatering phase results in the dry solid content of the sludge

remaining on the basin surface as sludge residue, whereas the majority of its water content continues to flow vertically through the sludge residue and basin filter. The water content in the sludge residue is further reduced through evapotranspiration. In addition to dewatering, the organic solids in the sludge are mineralized, thereby reducing organic solid in the sludge volume. The overall reduction of the sludge volume occurs without the use of any chemicals such as polymers. The process involves only a very low level of energy consumption for pumping the sludge and reject water (Nielsen 2003).

Sludge treatment reed bed systems

The feed sludge and sludge residue quality results presented in this study are from four different STRBs situated in Denmark: Nakskov (56° 49' 56 N; 12° 12' 00), Kallerup (55° 39' 56 N; 12° 12' 4 E), Helsingø (56° 44' 53 N; 12° 11' 40 E) and Kolding (55° 28' 20 N; 9° 34' 27 E). The STRB systems in Nakskov, Kallerup, Helsingø and Kolding were established in 1990, 1996, 1996 and 1998, respectively.

The four systems built by Orbicon all have the same design and construction and have all been planted with common reeds (*Phragmites australis*). The systems are dimensioned for a treatment capacity of 50–60 kg DS/m²/year and have received sludge from wastewater treatment plants (WWTPs) designed with mechanical, biological and chemical treatment. The number of basins in the four STRB systems are 8, 10, 10 (four new basins in operation from 2014) and 13, and the total filter areas are 4960, 9920, 10,500 and 36,530 m², respectively, in Kallerup, Nakskov, Helsingø and Kolding. The dry solid content in the feed sludge in the four systems is 0.5–1.5 DS%.

For more details about the systems and feed sludge, see Nielsen et al. 2014 and Matamoros et al. 2012.

The climate in Denmark is temperate with cool summers and mild winters. The average annual temperature is 8.7 °C (1.0 °C in January and 17 °C in August), and the annual precipitation is 400–900 mm.

Sampling and analyses

The standard procedure for sampling of the sludge residue was as follows: Sampling sites were randomly selected within each basin of the STRBs, and a total of approximately 8–15 sludge subsamples from the sludge residue (depth of 1.0–1.3 m) were collected. Samples were collected by a stainless steel corer (typically with a diameter of 3 cm), and all samples from each basin were then pooled and mixed. Values presented in Table 4 are based on pooled samples (minimum one pooled sample per basin in each STRB system). All samples have been analyzed by an accredited laboratory following standardized methods (ISO 5667-1:2006(E)) listed in Table 2.

The samples of the sludge residue from the four systems have been collected just prior to emptying of the individual basins during the years 2007–2011 (Nakskov), 2003–2005 (Kallerup), 2005–2008 (Kolding) and 2005–2008 (1. Emptying period from all 10 basins) and 2011, 2012 and 2014 (2. Emptying period from 6 basins) (Helsingø). The sludge residue has had treatment periods of a minimum of 8 years up to 20 years.

Table 2 Danish standard operational procedures

Parameter	Method
Dry solid	DS 204
Loss of ignition	DS 204
Total nitrogen (TN)	NF 1975
Total phosphorus (TP)	DS259/SM3120ICP
Lead (Pb)	DS259/SM3120ICP
Cadmium (Cd)	DS259/SM3120ICP
Chrome (Cr)	DS259/SM3120ICP
Cobber (Cu)	DS259/SM3120ICP
Quicksilver (Hg)	SM3112AAS _{co.vap}
Nickel (Ni)	DS259/SM3120ICP
Zink (Zn)	DS259/SM3120ICP
Organic hazardous compounds	GC/MS/SIM AK.87
LAS	LC/UV AK.87
PAH (16)	GC/MS/SIM AK.87
NPE	GC/MS/SIM AK.87
DEHP	GC/MS/SIM AK.87

Results and discussion

Dewatered and stabilized sludge represents a valuable nitrogen and phosphorus fertilizer for agriculture and, moreover, if applied on agricultural lands, contribute to the overall national and European goal of increasing the proportion of recycling of our waste streams. However, as sludge contains pollutants which may be harmful to the environment and because of the huge quantities produced, there are strong focus on the regulations and legislations for sludge treatment, handling and application on both national and EU level. The final sludge quality applied to agriculture depends not only on the constituents in the wastewater but also to a high degree on the sludge treatment method.

Mineralization of sludge

During the treatment period, in the STRB, organic content in the sludge will be mineralized fast (days, weeks), while the more stable parts (e.g. polyaromatic carbohydrates, lignin) will degrade much more slowly (years) (Nielsen 2005; Peruzzi et al. 2013). Because of the mineralization of the sludge over the years, which is enhanced by aeration through cracks in the dewatered sludge, rhizomes and from the drainpipe system, the deepest and oldest sludge layers typically have the lowest concentration of organic solid and carbon content, when related to dry solid content (Nielsen et al. 2014).

The total mineralization of the sludge (full sludge profile) during the entire treatment period for the four STRB systems can be calculated to between 10 and 27 % (Table 3). The calculation is based on the organic solid content (loss on ignition (LOI) data) before and after treatment. The deeper (and oldest) layers are typically more mineralized than that of the upper layers. The relative large variation in the total mineralization percentage between the STRB systems can be due to many aspects, hereunder the content of fat and oil. Sewage sludge with high contents of fat and oil often has a lower draining ability thereby reducing the aeration and mineralization (Nielsen 2011, 2005).

Of the four STRBs, Kolding had the highest fat content in the sludge residue, which may be the reason why Kolding STRB also had a very low mineralization (Table 2), the lowest DS% content and the highest organic fraction (determined as LOI) in the sludge residue (Tables 2 and 4). In a previous paper (Nielsen 2011), it was found that despite a large number of basins, low area loading and long resting periods to dry out, it was not 'sufficient to ensure healthy reeds, proper dewatering and mineralization of the sludge, if the quality of the sludge has too high a content of organic compounds including fat or oil'.

Table 3 Average organic content expressed as loss on ignition (LOI%±standard deviation) in feed sludge and in the sludge residue prior to emptying. The mineralization percentage is calculated as the relative difference before (feed sludge) and after treatment (sludge residue)

STRB system	Feed sludge		Sludge residue		Mineralisation (%)
	Sampling period	LOI (%)	Sampling period	LOI (%)	
Helsinge ^a	2000–2007	52±9	2005–2008	46±2 ^a	12
Helsinge ^b	2000–2014	54±11	2011, 2012, 2014	41±13 ^b	24
Kolding	2000–2007	56±5	2005–2007	54±6	4
Nakskov	2006–2008	51±5	2006–2008	46±5	10
Kallerup	2001–2013	56±3	2003–2005	41±6	27
Average mineralization					18 ^c

^a 1st emptying, 2005–2008

^b 2nd emptying, 2011, 2012 and 2014

^c In the average mineralization percentage, Kolding STRB is excluded due to the feed sludge to the STRB system that consisted of a mixture of anaerobic digested sludge and activated sludge contrary the three other systems

Sludge residue quality

Table 4 shows the sludge residue quality prior to emptying for four STRB systems. It can be seen that the sludge has obtained

high dry solid contents of on average 22 %. Moreover, all values, except for mercury and cadmium, are well below the Danish permissible values, which as mentioned are among the strictest in the EU. When based on ‘mg/kg DS’, mercury and

Table 4 Sludge residue quality parameters prior to emptying (±standard deviation) and Danish legal limits. The data presented are average values based on 8–10 pooled samples (approx. one pooled sample per basin in each STRB system)

Sludge residue after final treatment							
STRB	Helsinge (1st)	Helsinge (2nd)	Nakskov	Kallerup	Kolding	Average	Danish limits
Start of basins (year)	1996	1996	1990	1996	1998		
Basins emptied (year)	2005–2007	2011–2014	2007–2011	2003–2005	2005–2008		BEK No. 1650 of 13 December 2006
Dry solid (%)	22±4	26±10	21±4	24±6	19±3	22	
LOI (%)	46±2	41±13	48±4	41±6	54±5	46	
Total N (mg/kg DS)	26,300±4752	16,214±9471	30,000±3480	33,667±2338	35,650±6264	28,366	
Total P (mg/kg DS)	33,889±2328	25,983±9369	40,375±2441	37,000±2966	41,300±6010	35,709	
Pb (mg/kg DS)	52±9	28±9	79±12	65±8	64±14	57	120
Pb (mg/kg TP)	1529±193	1288±223	1914±268	1800±335	1554±367	1617	10,000
Cd (mg/kg DS)	1.2±0.2	0.5±0.1	1.7±0.3	1.1±0.4	1.7±0.3	1.2	0.8
Cd (mg/kg TP)	36±7	22±6	43±6	29±10	41±4	34	100
Cr (mg/kg DS)	31±9	19.0±6	34±5	28±3	35±8	29	100
Cu (mg/kg DS)	372±20	270±120	233±33	323±19	419±154	323	1000
Hg (mg/kg DS)	0.7±0.2	0.6±0.2	2.3±0.7	0.8±0.2	1.1±0.5	1.1	0.8
Hg (mg/kg TP)	20±5	28±6	56±18	23±5	31±16	31	200
Ni (mg/kg DS)	22±2	19±7	32±4	37±4	26±4	27	30
Ni (mg/kg TP)	578±180	862±78	784±86	1015±166	632±62	774	2500
Zn (mg/kg DS)	627±40	456±200	1140±153	880±92	929±179	806	4000
Oil (mg/kg DS)	88±86	91±62	55±247	220	1355±1822	362	
Fat (mg/kg DS)	335±563	2321±3968	209±517	1500	4643±6696	2790	
DEHP (mg/kg DS)	2.7±0.9	1.6±0.6	4.8±1.7	5.8±2.0	15±9.0	6	50
NPE (mg/kg DS)	0.8±0.3	0.6±0.1	0.5±0.6	3.0±0.8	8±4	2.5	10
PAH (mg/kg DS)	0.9±0.5	0.5±0.3	2.5±0.3	1.1±0.2	1.9±0.7	1.4	3
LAS (mg/kg DS)	<50	<50	<50	57±19	59±16	53	1300

LOI loss of ignition

cadmium exceed the Danish permissible values; however, when related to phosphorous (mg/kg TP), the concentrations are below the legal limits.

Heavy metals

Of the four systems investigated, zinc, Ni and Pb were the predominant heavy metals in the sludge, which also has been observed in other reed bed studies (Peruzzi et al. 2011; Uggetti et al. 2010). Heavy metals are mainly bound to particles in the sludge residue, and the translocation of heavy metals out of the systems with the drainage is therefore limited (Nielsen et al. 2014; Kolečka et al. 2008; Matamoros et al. 2012).

The mineralization of organic solid, leaving the heavy metals behind, may push the heavy metal levels in the oldest layers to concentrations higher than the legal standards for soil application (BEK No. 1650 of 13 December 2006) when expressed as milligrammes pre kilogramme DS (Matamoros et al. 2012) depending on the quality of the feed sludge. However, the quality of the sludge residue in general and even for sludge residues older than 20 years complies with the Danish regulations when based on the contents of heavy metals in relation to phosphorus. This is due to the fact that the phosphorus concentration in the sludge residue tends to increase in concert with heavy metals as a consequence of organic solid mineralization.

Nutrients

The nitrogen and phosphorus content in the sludge residue is typically high and the reason why sludge is valued as a fertilizer (Kolečka and Obarska-Pempkowiak 2013). Nitrogen typically decreases during the treatment period and with the depth of the sludge. For instance, in Nakskov STRB, it decreased by more than 45 % from 33.5 g/kg DS in the top layer (0–10 cm) to 18.4 g/kg DS in the bottom layer (120–130 cm) over a treatment period of 20 years for the oldest layer (Matamoros et al. 2012). This reduction is caused by microbial mediated processes of nitrification and denitrification as described by Nielsen et al. (2014). In addition, some nitrogen leave the system with the drainage (reject water) mainly in the form of nitrate and are returned to the wastewater treatment plant. Total phosphorous content tended to increase in general for the whole sludge residue and even with depth. Nakskov STRB confirms experience from other systems, where the long-term stabilization of domestic sludge causes an increase in the phosphorous concentration (Matamoros et al. 2012). Part of the phosphorous interacts with iron and other constituents in the sludge and is thus bound in the sludge residue matrix.

The application rate of sludge on farmland is, as mentioned, determined by the phosphorous and nitrogen content

in the sludge. The maximum allowable amount of phosphorous is 90 kg TP/ha every third year, and for nitrogen, it is 170 kg TN/ha/year (Danish legislation for sludge, BEK No. 1650 of 13/12/2006). On average TP and TN values in the sludge residue for the four STRB systems were 35 and 28 g/kg DS, respectively (Table 4). When related to the legal P application limit, this corresponds to approx. 2.6 t DS sludge per hectare, which can be spread on agricultural land every third year without exceeding the P limit (if related to TN, 3.2 t DS/ha could be applied; the lowest amount sets the limit).

Hazardous organic compounds

Studies show that hazardous organic compounds also are mineralized during treatment in STRB (Nielsen 2005; Federle and Itrich 1997). In a study of the mineralization of LAS and NPE in digested sludge treated in a STRB, a degradation of 98 % of LAS and 93 % of NPE was observed under aerobic conditions (Nielsen 2005; Danish Environmental Agency: Report no. 22, year 2000). The study demonstrated that oxygen was the limiting factor in the degradation of organic pollutants, while temperature has influence on the rate of degradation. Oxygen influx into the sludge improved the mineralization of LAS and NPE considerably, while mineralization under anaerobic conditions was very limited. In the same study, reductions of approx. 60 and 32 % were obtained for DEHP and PAH, respectively. The organic pollutants were mineralized in the whole depth and not only in the upper layer of sludge. While LAS, NPE, DEHP and PAH in storage experiments with anaerobically digested sludge (representing mechanical treatment) only were partly degraded in the top layer (0–20 cm) and below 20 cm, there was no degradation. Degradation under anaerobic conditions was negligible, which is comparable with the known knowledge regarding hazardous organic compounds from literature (Ejlertsson et al. 1996; Leahy and Colwell 1990; Marcomini et al. 1989; Marcomini et al. 1991).

Conclusions

Based on experience from Danish STRBs, the technology has proven to be an efficient methodology for the dewatering of sludge. In conclusion, the final sludge residue after a 10- to 20-year treatment period from the four Danish STRB systems complies with the limits put up in both Danish and EU legislation for sludge. The sludge residue has a quality, which makes it suitable for land application. The content of nutrition is high, so the sludge can be used as a substitute for commercial fertilizer. The quality of the final product in the STRB with the low content of heavy metals and hazardous organic

compounds makes it possible to recycle the sludge residue to agriculture, too. In conclusion, the STRB is, if the system has the right dimension, design and operation in relation to the sludge quality, a feasible technology for the sludge treatment and stabilization before its final land disposal.

Acknowledgments The authors want to thank the technical staff at the Waste Water Treatment Works for their technical assistance during the operations and sampling.

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