




Towards carbon neutrality and circular economy: an innovative combination of enhanced biogas production and nutrient recovery from sludge dewatering liquor at a municipal wastewater treatment plant in Germany

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ABSTRACT

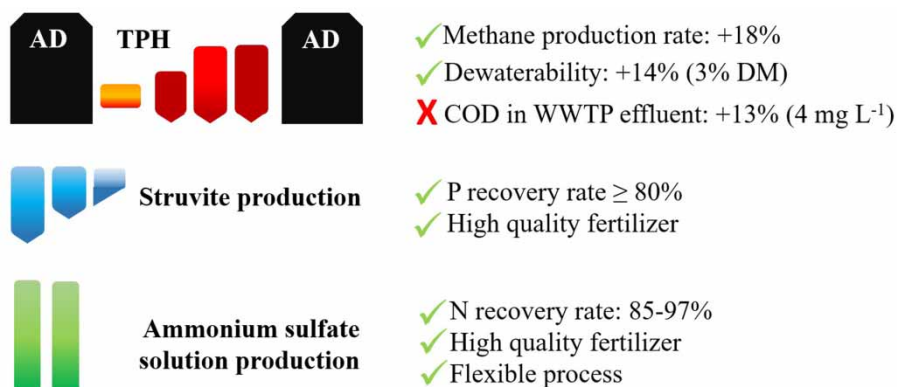
An innovative circular economy (CE) system was implemented at the wastewater treatment plant (WWTP) in Brunswick. The performance of the CE system was evaluated for 4 years: the thermal pressure hydrolysis enhanced the methane production by 18% and increased the digestate dewaterability by 14%. Refractory COD formed in thermal hydrolysis and increased the COD concentration in the WWTP effluent by 4 mg L⁻¹ while still complying with the legal threshold. Struvite production reached high phosphorus recovery rates of >80% with a Mg:P molar ratio ≥0.8. Nitrogen was successfully recovered as ammonium sulfate with high recovery rates of 85–97%. The chemical analyses of secondary fertilizers showed a low pollutant content, posing low risks to soil and groundwater ecosystems. The total carbon footprint of the WWTP decreased due to enhanced biogas production, the recovery of renewable fertilizers and a further reduction of nitrous oxide emissions. Using green energy will be crucial to reach carbon neutrality for the entire WWTP.

Key words: ammonium sulfate solution, biogas, carbon footprint, circular economy, energy recovery, struvite, thermal pressure hydrolysis

HIGHLIGHTS

- Full-scale implementation of a circular economy approach at a municipal WWTP.
- 18% increase in methane production due to thermal pressure hydrolysis.
- High recovery of struvite and ammonium sulfate from sludge liquor.
- High quality of secondary fertilizers with low pollutant concentration.
- Decrease in carbon footprint due to energy and fertilizer recovery and lower nitrous oxide emissions.

GRAPHICAL ABSTRACT



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ABBREVIATIONS

AD	anaerobic digestion
ASL	ammonium sulfate solution
CE	circular economy
CO ₂ e	CO ₂ equivalent
COD	chemical oxygen demand
CHP	combined heat and power
DLD	digestion-lysis-digestion
DM	dry matter
EU	European Union
GWP	global warming potential
ICP-OES	inductively coupled plasma optical emission spectroscopy
LCA	life cycle assessment
NH ₄ -N	ammonium indicated as nitrogen units
oDM	organic dry matter
PE	population equivalents
PO ₄ -P	phosphate indicated as phosphorus units
rCOD	refractory chemical oxygen demand
TKN	total kjeldahl nitrogen
TN	total nitrogen
TP	total phosphorus
TPH	thermal pressure hydrolysis
TSS	total suspended solids
WAS	waste-activated sludge
WWTP	wastewater treatment plant

1. INTRODUCTION

The European Union (EU) Green Deal aims to reorganize the EU economy for a sustainable future. Therefore, it focuses on different fields of action such as climate protection, renewable energy, a pollution-free environment and a circular economy (CE). A CE in the water sector can highly contribute to a pollutant-free environment and to the production of carbon neutral energy by reclaiming water and recovering nutrients/materials and energy from wastewater. In Germany, a CE system was implemented at the wastewater treatment plant (WWTP) in Brunswick. The WWTP was originally designed for 275,000 population equivalents (PE). However, nowadays the WWTP receives wastewater from 350,000 PE. To avoid an overload of the conventional activated sludge system and to comply with the legally required effluent quality, the plant owner decided to implement an innovative combination of three CE-related technologies. The innovative system removes phosphorus and nitrogen from the return load of the sludge liquor to the headworks of the WWTP. It consists of a thermal hydrolysis process to enhance the biodegradation in the subsequent digester and increase the ammonium and phosphate concentrations in the sludge liquor, a phosphorus recovery process producing struvite and a nitrogen recovery process to produce ammonium sulfate solution. Hereby, the benefit is that renewable fertilizers and energy in the form of biogas are produced and thus, allow to close the nutrient and energy loops.

Thermal hydrolysis processes are applied in sludge treatment to enhance its dewaterability, its solubilization and to reduce its viscosity (Barber 2016). In consequence, the biodegradability of the hydrolyzed sludge in anaerobic digestion is increased, leading to a higher methane yield (Neyens & Baeyens 2003; Zhen *et al.* 2017; Liu *et al.* 2019). Different types of thermal hydrolysis processes exist such as thermal alkaline hydrolysis (Toutian *et al.* 2020a) or the combination of a thermal high-pressure boiling and decompression. The latter will be called thermal pressure hydrolysis (TPH) in this publication. In the TPH, the digested sludge is 'cooked' at high temperatures and pressures which can range between 140 and 170 °C and 5 and 8 bars, correspondingly. The subsequent rapid decompression, when the sludge passes a small orifice, leads to high mechanical shear forces destroying microbial cell walls and releasing soluble organic compounds. Soluble non-biodegradable organic compounds also known as refractory COD can be formed, starting already at process temperatures of 130 °C, but especially at higher temperatures above 160 °C, increasing the COD concentration in the effluent of a WWTP (Toutian *et al.* 2020b). In addition, the release of nutrients such as ammonium and phosphate is also increased due to the enhanced

biodegradation in the second digester. Combining the process with a subsequent nutrient recovery unit, the nutrient release is an advantage.

Many different processes for phosphorus recovery either from ash, sludge or sludge dewatering liquor exist (Desmidt *et al.* 2015). Struvite is a magnesium ammonium phosphate compound ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) and is known as a slow-release fertilizer (Kratz *et al.* 2019) with all three nutrients being plant-available (Watson *et al.* 2019). Controlled struvite precipitation has the additional purpose of preventing the subsequent units from scaling and clogging due to undesired struvite precipitations, e.g. in the heat exchanger of a subsequent stripping process. A struvite production unit for phosphorus recovery from sludge dewatering liquor consists usually of several reactors for CO_2 stripping to increase the pH, struvite formation and settling of the struvite crystals. For struvite formation, NaOH and MgCl_2 are dosed to further increase the pH and to induce precipitation and crystallization, respectively. Park *et al.* (2020) and Rahman *et al.* (2014) report phosphorus recovery rates from sludge liquor between 80 and 90%. Hence, the phosphorus return load to a WWTP can be significantly decreased by applying this process.

At municipal wastewater treatment plants in Germany, nitrogen is typically removed via nitrification and denitrification. However, when a WWTP treats its primary and waste-activated sludge (WAS) via digestion, usually the organic nitrogen, contained in the sludge, is converted into ammonium during digestion increasing the nitrogen return load in the sludge liquor to the headworks of a WWTP. To decrease the nitrogen return load, a nitrogen recovery unit can be used. Nitrogen recovery via air stripping and scrubbing is a well-known and mature technology. The process is based on chemical reactions only and depends mainly on the pH and the temperature. The higher the pH and temperature are, the more the chemical equilibrium between ammonium and ammonia shifts to the ammonia side. Under these conditions, the ammonia can be stripped out with air. In a subsequent scrubber operated with sulfuric acid, the ammonia is converted back to ammonium due to the acidic pH and reacts together with the sulfuric acid to diammonium sulfate also known as ammonium sulfate solution. The effectiveness of recovered ammonium sulfate solution as a fertilizer was shown to be as good as that of a commercially available ammonium sulfate solution (Szymańska *et al.* 2019). Using the sludge dewatering liquor of the digestate to recover nitrogen, the nitrogen return load to the WWTP can be strongly decreased between 70 and 92% (Sengupta *et al.* 2015).

The life cycle assessment (LCA) is a method to assess and evaluate the carbon footprint of a treatment concept. Many LCA studies deal with either struvite recovery (Pedizzi *et al.* 2018; Lam *et al.* 2020) or with ammonium sulfate recovery (Kar *et al.* 2023; Saud *et al.* 2023), but to the author's best knowledge, there is no study published considering the synergies of a CE system comprising phosphorus, nitrogen and energy recovery.

This publication presents the first three years of operation of the CE technologies and includes the last year of operation with the old configuration for comparison, it evaluates the performance of the new CE solutions and characterizes the qualities of the renewable fertilizers. Furthermore, the carbon footprint of the CE solutions is assessed considering the synergies between the technologies and the crucial factors to achieve carbon neutrality of the wastewater treatment.

2. MATERIALS AND METHODS

2.1. WWTP in Brunswick with energy and nutrient recovery units

The municipal WWTP in Brunswick applies a primary treatment and the conventional activated sludge process to treat its wastewater (Figure 1). In the conventional activated sludge process, aerobic carbon degradation, nitrification, denitrification and enhanced biological phosphorus removal as well as chemical phosphorus removal are integrated. Half of the WWTP effluent is post-treated in infiltration fields and the other half is used for irrigation of agricultural fields in summer. Until 2019, the mixed sludge was treated in a one-stage digestion only, dewatered and incinerated. In the new configuration, a TPH was implemented between two digesters also known as digestion-lysis-digestion (DLD) configuration using the Haarslev process. Hereby, the WAS is treated in the first digestion stage, then dewatered and hydrolyzed, increasing the availability of ammonium and phosphate as well as enhancing the biogas production rate due to a higher biodegradability in the subsequent digestion stage. There, the hydrolyzed sludge is digested together with the primary sludge and the fat, oil and grease (FOG) fraction. The digestate is dewatered and will be incinerated in the long term during the entire year. However, currently, the digestate is still valorized as organic fertilizer in summer and used for fertigation together with the WWTP effluent. The agricultural reuse of the digestate will end in 2028 and was therefore not considered for the carbon footprint analyses. The filtrate from pre-dewatering of the digested WAS is used to recover phosphorus via struvite production and to recover ammonia via the production of ammonium sulfate solution using ammonia air stripping and scrubbing with sulfuric acid. In the near

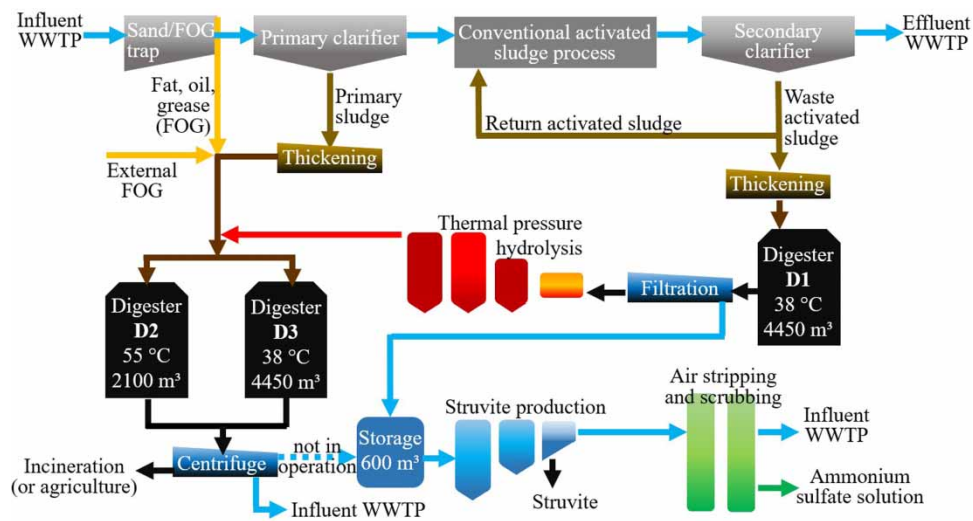


Figure 1 | Flow scheme of the WWTP in Brunswick including the new CE system: TPH, struvite production to recover phosphorus and air stripping and scrubbing to recover nitrogen as ammonium sulfate solution from sludge liquor.

future, the centrate from the second digestion stage will also be treated in the nutrient recovery plant, which is currently indicated as ‘not in operation’.

2.2. Operating parameters

The focus is put on the new CE system comprising the DLD system and the nutrient recovery units.

2.2.1. Anaerobic digestion

As shown in Figure 1 and Table 1, the first digester (D1) was operated at 38 °C and received WAS with an organic loading rate of around $2 \text{ kg oDM (m}^3 \text{ d)}^{-1}$. The other two digesters received FOG, primary sludge and hydrolyzed WAS. The smaller digester (D2) was operated at 55 °C and the larger digester (D3) at 38 °C. The organic loading rates were around 4.4 and $2.7 \text{ kg oDM (m}^3 \text{ d)}^{-1}$, respectively. The hydraulic retention times were around 17 days for D1 and on average for D2 and D3 around 15 days. The degradation rates were 40 and 48% for D1 and D2/D3, respectively. The nutrient concentrations in the digestate of D1 were $2,400 \text{ mg P L}^{-1}$, $450 \text{ mg PO}_4\text{-P L}^{-1}$ and $1,500 \text{ mg NH}_4\text{-N L}^{-1}$, while the nutrient concentrations in the mixture of the digestates resulting from D2/D3 were $1,900 \text{ mg P L}^{-1}$, $310 \text{ mg PO}_4\text{-P L}^{-1}$ and $1,200 \text{ mg NH}_4\text{-N L}^{-1}$.

2.2.2. TPH (Haarslev)

Before hydrolyzing the digested WAS and after the addition of a polymer, the digestate was dewatered/filtered in a screw press to a DM content of 13%. The thickened sludge was hydrolyzed in the TPH at temperatures mainly between 150 and

Table 1 | Average operational parameters of digesters, when TPH is in operation

Parameters	D1	D2	D3
Volume (m ³)	4,450	2,100	4,450
Temperature (°C)	38	55	38
Organic loading rate (kg oDM m ⁻³ d ⁻¹)	2	4.4	2.7
Hydraulic retention time (d)	~17		~15
Volatile solids reduction rate (%)	40		48
TP concentration (mg L ⁻¹)	2,400		1,900
PO ₄ -P concentration (mg L ⁻¹)	450		310
NH ₄ -N concentration (mg L ⁻¹)	1,500		1,200

160 °C and a pressure of around 6 bar. The hydraulic retention time in the TPH reactor was usually 90 min. The hydrolyzed sludge had usually a temperature of 105 °C. It was mixed with thickened primary sludge and FOG, having a temperature of around 15 °C, resulting in a lower temperature than that of the subsequent second digestion stage, where it was sent afterwards.

2.2.3. Struvite production unit (NuReSys)

The NuReSys process produces struvite from sludge dewatering liquor. In the investigated period, the struvite production unit received only the filtrate from the first digestion stage. However, for the carbon footprint of the CE scenario both, the filtrate and centrate were considered. The total suspended solids (TSS) content was around 300 mg L⁻¹, below 600 mg L⁻¹ as required to avoid contamination with e.g. organic particles. The NuReSys system consists of three reactors. To increase the pH to >8, CO₂ was stripped in the first reactor. In the struvite reactor, NaOH was added to increase further the pH and MgCl₂ was dosed to induce struvite precipitation. Hereby, the Mg:P molar ratio was maintained mainly between 0.5 and 1.5.

2.2.4. Air stripping and scrubbing to produce ammonium sulfate solution (RVT process equipment)

The operating temperature in the air stripping unit was maintained between 55 and 65 °C and the pH was elevated mainly between 9.5 and 11 by dosing NaOH to the sludge liquor. To prevent the heat exchangers from scaling and clogging, the TSS content in the liquor was kept below 600 mg L⁻¹ by separating the struvite crystals successfully from the sludge liquor in the settling tank and using a hydrocyclone.

2.3. Laboratory analyses

Sludge liquor samples were analyzed using test kits from Hach for the chemical oxygen demand and phosphate. For sludge liquor and ammonium sulfate solution samples, the inductively coupled plasma optical emission spectroscopy (ICP-OES) was used to determine calcium (Ca), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), magnesium (Mg), nickel (Ni), potassium (K), total phosphorus (TP) and zinc (Zn) (DIN EN ISO 11885:2009-09-E22). Ammonium (NH₄-N), mercury (Hg), total Kjeldahl nitrogen (TKN) and total nitrogen (TN) were analyzed according to DIN 38406:1983-10-E5-2, DIN EN 1483:2007-07 E12-Abs. 4, DIN EN 25663:1993-11-H1 and DIN ENV 12260:2003-12(H43), respectively. Struvite samples were analyzed using the aqua regia digestion method in combination with the ICP-OES to determine the contents of aluminum (Al), arsenic (As), boron (B), Cd, Ca, Cr, cobalt (Co), Cu, iron (Fe), Pb, Mg, manganese (Mn), Ni, K, sulfur (S), thallium (Tl), TP and Zn (DIN EN ISO 22036:2009-06). The contents of TN and Hg were analyzed as described in VDLUFA A2.2.3: 1991 and DIN ISO 16772:2005-06, respectively. The TSS, dry matter (DM) and organic dry matter (oDM) contents were determined using DIN 38414-S2:1985-11 and DIN 38409-H1-1/-3:1987:01.

2.4. Carbon footprint assessment

2.4.1. Goal and scope definition

The goal of the CO₂e-footprint was to analyze the potential environmental impacts of the WWTP with the new CE system compared to the situation before. The function of the system is 'to provide wastewater treatment according to the legal requirements' including all processes related to this function. The functional unit was defined via the annual organic load of the WWTP calculated per PE and year of the WWTP. The CO₂e-footprint analyzed the entire system of wastewater and sludge treatment and disposal. Therefore, the system boundary included the entire WWTP including the agricultural reuse of water. Water and nutrients delivered to agriculture were accounted as credit equivalent for avoiding mineral fertilizers and pumping groundwater. Potential field emissions of nutrient reuse were accounted as well as avoided emissions from mineral fertilizers. Biogas was utilized in a combined heat and power unit (CHP) and electricity was accounted. Heat was reused in the WWTP as far as possible, however, excess heat in the summer months was not accounted as credit. This non-utilized heat amount is about 25% of the annual heat production of the CHP (Kim *et al.* 2023). In the carbon footprint assessment both sludge liquors, from the screw press and the centrifuge were assumed to be united as it is planned for the near future. For the calculation of the CO₂e-footprint, the global warming potential (GWP) for a time horizon of 100 a (IPCC 2014) was used. The WWTP was modeled with the LCA and flow modeling software Umberto LCA+ (ifu 2019) using the EcoInvent database v3.6 (EcoInvent 2021). The used datasets in terms of background data are summarized in Table S1 (supplementary material).

2.4.2. Input data for LCA

The inventory data for the CO₂e-footprint were based on the operational data presented in this publication. The sludge and sludge liquor quantities and qualities for the baseline and CE scenarios are presented in the supplementary material in detail (Table S2 and Table S3). Within the baseline scenario, primary and WAS are digested together (28 kWh_{th} m⁻³, yield: 0.26 m³ CH₄ (kg oDM)⁻¹) and the digested sludge is dewatered with a centrifuge to 22% DM.

In the CE scenario, WAS is separately digested (28 kWh_{th} m⁻³, yield: 0.22 m³ CH₄ (kg oDM)⁻¹). Afterwards, the digested WAS is pre-dewatered in a screw-press to 13% DM. The pre-dewatered digested WAS is hydrolyzed in a TPH (1.8 kWh_{el} m⁻³, 130 kg steam m⁻³ produced in a steam generator using biogas). Hydrolyzed hot WAS is mixed with primary sludge and FOG (15 °C) and digested again (7 kWh_{th} m⁻³, 0.26 m³ CH₄ (kg oDM)⁻¹). Digested sludge is then dewatered to 25% DM in a centrifuge.

The struvite reactor consumes 1.3 kWh_{el} m⁻³. Caustic soda dosing ranged mainly between 0.3 and 0.7 L NaOH (50%) m⁻³ corresponding to a P recovery rate of 80%. For LCA we assumed a P recovery rate of 95%, resulting in a caustic soda dosing rate of 1 L NaOH (50%) m⁻³. In operation the molar ratio for Mg:P ranged mainly between 0.5 and 1.5. For LCA, a molar ratio for Mg:P of 1.3 (equal to 2.3 L MgCl₂ (25%) m⁻³) was chosen, corresponding to a high precipitation and harvesting efficiency of 95%.

The ammonia stripper consumes 1 kWh_{el} m⁻³ and 9 kWh_{th} m⁻³. The NaOH dosages ranged from 2 to 6 L NaOH (50%) m⁻³ at different process temperatures, while for LCA 3.3 L NaOH (50%) m⁻³ and 60 °C were chosen. The scrubber consumes about 1.4 L H₂SO₄ (96%) (m³ sludge liquor)⁻¹. The efficiency for ammonium removal is also about 95%. All relevant numbers for the baseline scenario and the CE scenario are summarized in the supplementary material (Table S4). The annual inventory data including electricity demands of the entire WWTP are shown in detail in the supplementary material (Table S5).

3. RESULTS AND DISCUSSION

Four years of plant operation are presented in this chapter. During this time, the TPH and the nutrient recovery system were not always in operation, so it was possible to compare the effect with and without the new CE system in operation. Therefore, after a restart or shut-down of the CE system, the lag time for the effect of the TPH on the dewaterability of the digestate and on the concentration of the dissolved COD in the WWTP effluent was excluded from the data set. The lag times were estimated to 15 and 17 days.

3.1. Performance of the CE system

3.1.1. Thermal pressure hydrolysis

Figure 2 shows the first 300 days before the implementation of TPH as a reference of the process conditions without the new plant configuration. In the 3 years after its implementation (day 300–day 1,500), the TPH was not in continuous operation due to technical challenges such as broken elements or unexpected gas leakages, as indicated in Figure 2 (TPH: off and TPH: on). This led to the opportunity to observe and compare time periods, in which the digesters were operated with and without hydrolyzed WAS. In order to assess the performance of energy recovery, the methane production rate was chosen as a crucial parameter, because it combines both the gas production rate and the methane content. Without TPH in operation, the methane production rate ranged mainly between 230 and 290 m³ h⁻¹. TPH increased the methane production rate to a range between 280 and 330 m³ h⁻¹. The thermal process led to an increase in the methane production rate of 18% in its median (Figure 2(a)). Thereby, 9% of the entire biogas produced in the WWTP was needed for steam generation to run the TPH. The remaining additional biogas was utilized in the CHP. This resulted in a higher annual self-supply with heat and the usage of natural gas in winter could be reduced by approximately factor 3 (see Table S5, supplementary material).

The results of the TPH performance confirm the TPH effects shown by Neyens & Baeyens (2003) as well as by Razavi *et al.* (2021). Razavi *et al.* (2021) even observed an increase of 40% in their methane yield. According to the German network of experts for water, wastewater and waste (DWA 2014), the enhancement of the biogas production rate of 15–25% is a typical consequence of the thermal hydrolysis process. Also, Toutian *et al.* 2020b showed increases in their biomethane potential between 17 and 27% for thermal hydrolysis process temperatures between 130 and 170 °C. This corresponds to the observed enhancement in Brunswick.

The increased methane production rate results from the higher substrate availability originating from soluble organic compounds formed during the TPH. Due to the breakdown of complex organic compounds and cell structures through the TPH

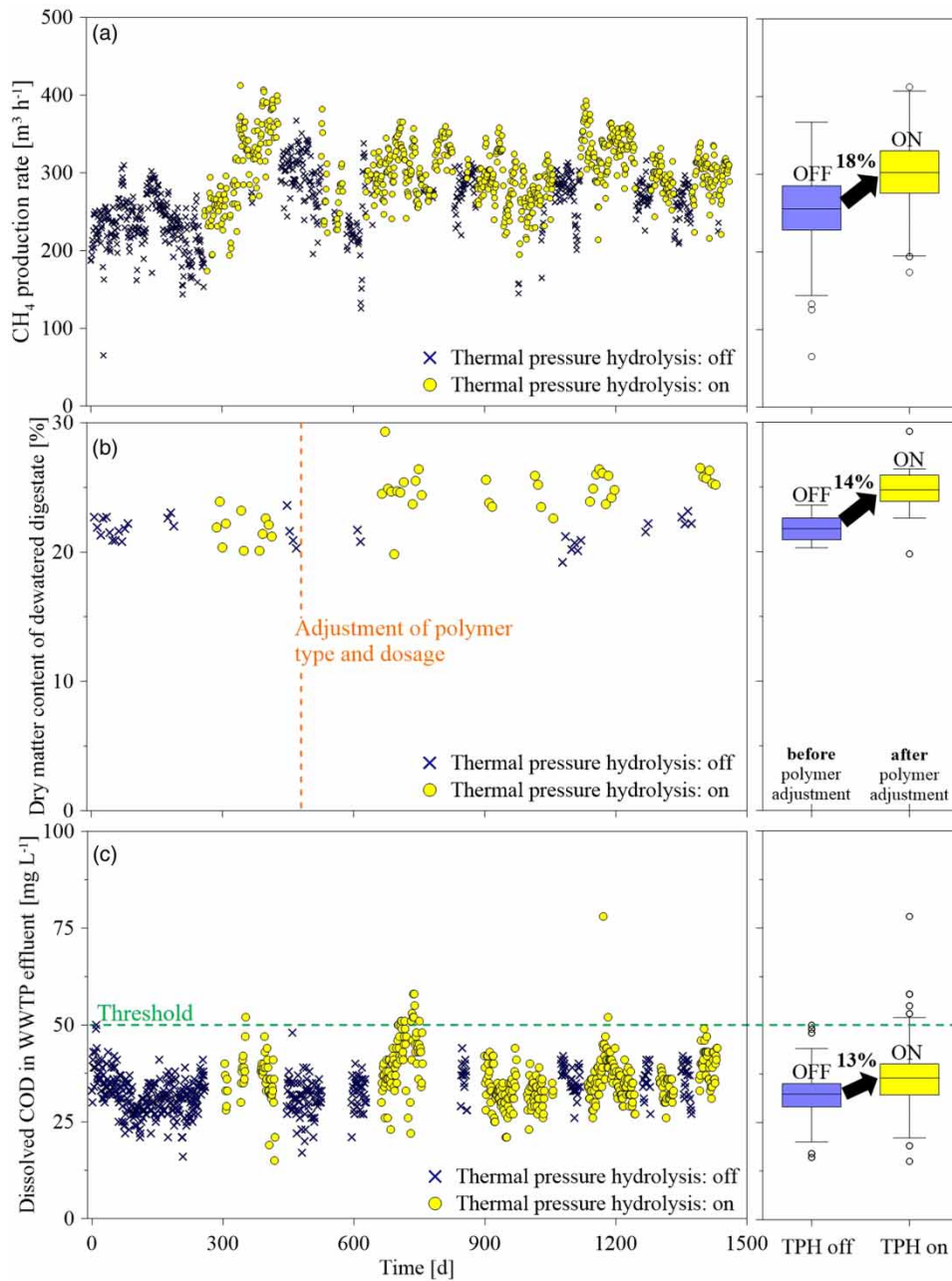


Figure 2 | (a) Methane production rate over 3 years of plant operation with (●) and without (×) hydrolyzed WAS as substrate: increase in production rate by a factor of 1.18. (b) DM content of dewatered digestate showing cleaned data: when TPH was longer in operation than the hydraulic retention time of ~15 days between TPH and centrifuge (c) Dissolved COD in WWTP effluent showing cleaned data: when TPH was longer in operation than the hydraulic retention time of ~17 days between TPH and WWTP effluent.

(Barber 2016), additional ammonium and phosphate can be released in the subsequent digestion stage. The increased concentrations of ammonium and phosphate are an important synergy for the nutrient recovery units. They will allow for a higher nutrient recovery efficiency referring to the entire WWTP due to the pretreatment with the TPH and the subsequent digestion stage as soon as the centrate from the second digestion stage will also be treated in the nutrient recovery unit.

Figure 2(b) shows the DM content of the dewatered digestate resulting from the second digestion stage operated with and without the addition of hydrolyzed sludge. As shown in Figure 2(b), after 500 days, the polymer type and dosage rate were adjusted, because of the changed sludge structure due to the TPH treatment, resulting in an increase in the DM content

after centrifugation of the digestate. To compare the effect of the TPH, the DM content before the adjustment and without hydrolyzed digestate was compared to the DM content after the adjustment containing hydrolyzed digestate. The TPH together with the adjusted polymer increased the median of the DM content from 22 to 25% DM by a factor of 1.14 and enhanced its dewaterability successfully.

Neyens & Baeyens (2003) and Zhen *et al.* (2017) reported also the enhancement of digestate's dewaterability after a thermal hydrolysis treatment. Barber (2016) summarized in his review an absolute increase of 10% DM with final DM contents ranging between 25 and 40% DM. In our study, only the WAS was thermally hydrolyzed and thus, the primary sludge and FOG did not contribute to the enhancement resulting in a lower final DM content. Still, our final DM content of 25% is in the range of the authors reviewed by Barber (2016), even though the absolute increase in the DM content of 3% DM is comparably low also in comparison to DWA (2014) and Zikakis *et al.* 2017, who indicated absolute increases of 8 and 9% DM. Nevertheless, our relative increase in dewaterability of 14% is still in the range of 12 and 30% revealed by Toutian *et al.* 2020b.

A drawback of thermal hydrolysis is the risk of the formation of soluble non-biodegradable organic compounds also known as refractory chemical oxygen demand (rCOD). Unfortunately, the rCOD was not determined. However, as a hint for the formation of rCOD, the dissolved COD in the effluent of the WWTP was investigated (Figure 2(c)). Comparing the dissolved COD concentration during TPH operation to that without the TPH in operation, its median increased from 32 to 36 mg L⁻¹ by a factor of 1.13. Similar observations were also made by Toutian *et al.* (2020b) who predicted based on their laboratory investigations an increase in the dissolved COD concentration in the WWTP's effluent between 2 and 15% for process temperatures between 130 and 160 °C.

3.1.2. Nutrient recovery units

3.1.2.1. Struvite production unit. The formation of struvite was induced by the addition of MgCl₂ and the struvite was harvested from the reactor so that the concentrations of phosphate (PO₄-P) and phosphorus (TP) in the outlet of the reactor decreased correspondingly. This decrease referred to its initial concentration was defined as recovery rate. The recovery rates were referred to two different parameters, the PO₄-P concentration and the TP concentration as shown in Figure 3.

Both, the PO₄-P-related recovery rate and the TP-related recovery rate varied highly within the first three years due to operational problems. The struvite crystals were so small (<1 mm), that a part of them were washed out and reached the subsequent ammonia recovery unit, clogging its heat exchangers. This led to lower recovery rates than expected. Several measures were undertaken to increase the struvite crystal size: The flow rate in the hydrocyclone was increased to better separate the fine crystals from the sludge liquor before leaving the struvite production unit. The settler's geometry was adapted by increasing the slope of its declining wall to allow for a better settling of the fine crystals. Delayed harvestings allowed for a longer retention time and, thus, a longer growth time of the crystals, but at the same time, a clogging event of the settler occurred. Since day 900, an enlarged stirrer length and a higher agitation speed were used to optimize the mixing conditions and small harvested crystals were reinjected using new nozzles to avoid any clogging. From day 1,100 on, during almost 100 days both recovery rates reached their expected range at and above 80%. Until then, the pH was maintained at around 8.5. Around day 1,200, the recovery rates started to vary again at a pH of 7.9. At this time, a power failure caused a reset of the control system operating at suboptimal conditions. The medians of the recovery rate related to the TP and of that referred to the PO₄-P reached 73 and 80%, respectively.

The PO₄-P-related recovery rate indicates the performance of the precipitation process when the dissolved PO₄-P is successfully converted into the solid PO₄-P compound of struvite. Compared to the recovery rate relating to the TP, the PO₄-P recovery rate was 9% higher. This corresponds to the difference in the inflow concentrations between TP and PO₄-P. The influent TP and PO₄-P concentrations varied mainly between 270 and 400 mg TP L⁻¹ and 250 and 350 mg PO₄-P L⁻¹, respectively. Hereby, the median value of the TP concentration was 325 mg TP L⁻¹ 12% higher compared to that of PO₄-P with 285 mg L⁻¹. Hence, the inflow contained also other phosphorus compounds than PO₄-P, which could not have been removed via the struvite precipitation and harvesting processes. In addition, again, especially in the first three years, fine struvite crystals were washed out and also contributed to a lower TP recovery rate compared to the PO₄-P recovery rate. Besides the pH, the availability of ammonium and magnesium is crucial for a successful precipitation process. The ammonium (NH₄-N) concentration varied between 1,100 and 1,400 mg NH₄-N L⁻¹. The molar ratio of N:P in the influent to the struvite reactor ranged mainly between 6 and 12. The molar ratio of Mg:P varied highly between 0.25 and 2 as shown in Figure 3(c). Only the

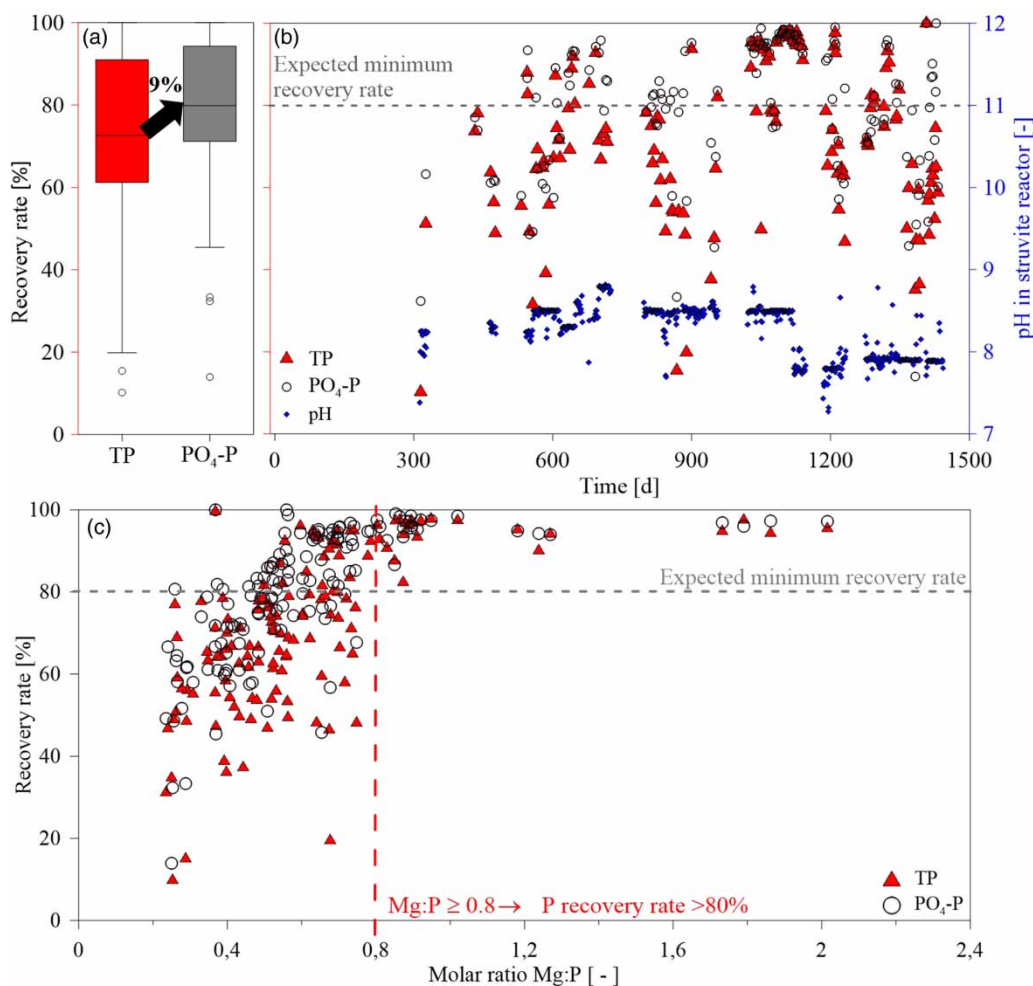


Figure 3 | (a) Total phosphorus (TP) and phosphate (PO₄-P) recovery rates over time and (b) TP and PO₄-P recovery rates depending on the molar ratio of Mg:P.

operation of the process at a molar ratio of 0.8 and greater led to a reliable recovery rate of 80% and higher. Hence, the optimal conditions for the struvite formation process were maintaining the pH at 8.5 together with a Mg:P molar ratio of 0.8.

This was also observed by Shaddel *et al.* 2019. They indicated a pH of 8.5 as optimum, with which they reached recovery rates of 80% and higher. Simultaneously, they maintained the Mg: N:P ratio between 1:2:1 and 1.67:12:1. Hence, their Mg:P molar ratio of 1 and 1.67 was even higher than in our study. Their N:P molar ratios between 2 and 12 were in a similar range as in our study. Thus, in our case, NH₄-N was not the limiting factor for struvite precipitation. Moreover, the low pH of 7.9 in combination with a too low Mg:P molar ratio and the operational problems mentioned before were the cause for low recovery rates.

Similar observations were made by Park *et al.* 2020. They reached high recovery rates between 83 and 91% at a pH between 8.25 and 8.5 and Mg:P molar ratios between 0.6 and 1.5. The comparison of 16 struvite production plants revealed also high recovery rates between 81 and 99% at pH values between 8 and 11 and Mg:P molar ratios between 0.8 and 1.6 (Rahman *et al.* 2014). In our case, high recovery rates of 97% were reached at a pH of 7.8, only when the Mg:P molar ratio was maintained at 2. This also corresponds to the results of Rahman *et al.* (2014). They reported also high recovery rates at a pH between 7 and 8 with Mg:P molar ratios between 1.2 and 2.4.

The recovery of PO₄-P decreased the phosphorus return load to the headworks of the WWTP for retreatment. Before the implementation of the struvite recovery plant, the yearly average TP concentration in the effluent was 0.7 mg L⁻¹. With the struvite recovery plant in operation, the yearly average TP concentration decreased to 0.6 mg L⁻¹ even though the plant was

not in permanent operation. Consequently, with the new plant configuration, the legal requirement of an effluent TP concentration below 1 mg L^{-1} was easily fulfilled (AbwV 2022). This will be even further enhanced, when the centrate from the second digestion stage will be treated together with the filtrate in the struvite recovery unit. In this case, referring to the TP influent load to the entire WWTP, the results correspond to a recovery rate of 16% under optimal and permanent operating conditions.

3.1.2.2. Ammonium sulfate production unit. Figure 4 shows the recovery rate referred to the ammonium and TKN influent concentrations to the air stripping unit. The influent TKN and ammonium concentrations ranged between 440 and $1,600 \text{ mg L}^{-1}$ and 570 and $1,500 \text{ mg L}^{-1}$, respectively. Both were on a similar level resulting in similar recovery rates. Within the first 200 days, the process temperature was at 55°C and the pH at 11.

From day 600 on, the temperature was increased to 65°C and the pH was decreased to pH 9.5. For both configurations, the recovery rates ranged mainly between 90 and 97% reaching their desired range. After day 1,000, the operating temperature was further decreased ranging between 50 and 55°C at a pH of 9.5. Those operating conditions delivered recovery rates below 85% which were considered as too low. Hence, the optimal operating conditions were at the temperatures of 55 and 65°C and at pH values of 11 and 9.5, respectively. The advantage of the operating conditions at the higher temperature and the lower pH is, that roughly 30% of NaOH could be saved.

Figure 4 also shows that the process can be switched on and off indicated by the high variation in pH and temperature and still delivers the desired recovery rates in the short term. The recovery rate referred to TKN and $\text{NH}_4\text{-N}$ was for both parameters roughly in the same range at 90 and 94%, respectively. After day 1,100, the operation of the unit was stopped, because the nitrogen concentration in the effluent of the WWTP complied with the legal threshold and hence, there was no demand to reduce the nitrogen return load to the WWTP. The reasons therefore are the currently still ongoing valorization of the digestate in agriculture for fertigation, resulting in a lower nitrogen return load, and the higher temperatures in summer contributing to a higher nitrogen removal rate in the conventional activated sludge system. The yearly average concentration of TN in the effluent of the WWTP before the implementation of the nitrogen recovery unit was 12 mg L^{-1} , which corresponded to the threshold defined by the local authority. It should be noted that the WWTP effluent in Brunswick is either reused in agriculture for fertigation or post-treated in infiltration fields so that the nitrogen load is further reduced until it reaches a water body. Since the implementation of the nitrogen recovery unit and further optimization of the conventional activated sludge process, the yearly average concentration of the TN in the WWTP effluent decreased to 9.3 mg L^{-1} complying with the legal requirements. This will be further enhanced in the near future when the centrate from the second digestion stage will also be treated in the nutrient recovery plant and hence, more nitrogen can be recovered. In this case, referring to the nitrogen load of the entire WWTP, our results correspond to a nitrogen recovery rate of 13% resulting from ammonium sulfate solution and struvite production for a permanent operation and under optimal conditions.

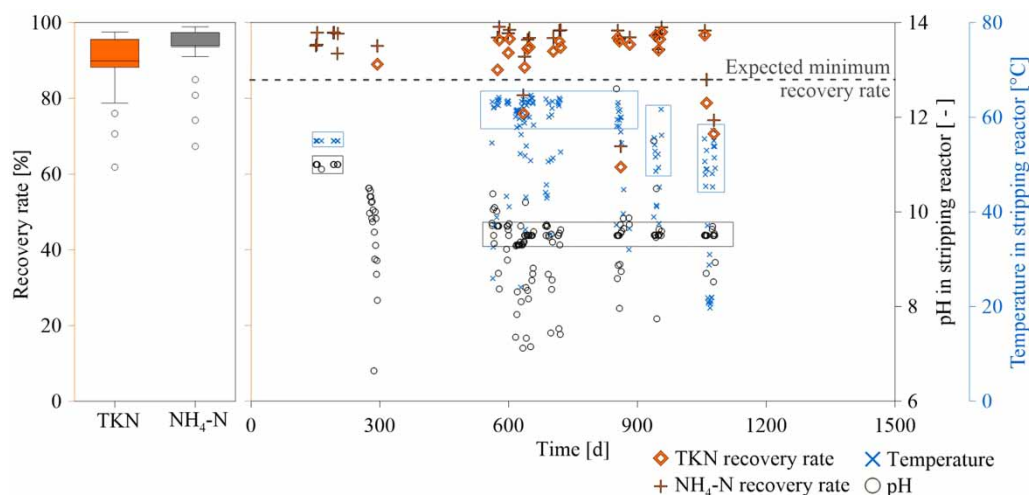


Figure 4 | (a) Nitrogen recovery rates measured as TKN and $\text{NH}_4\text{-N}$ as box plots and (b) both recovery rates depending on the time with pH and temperature conditions in the stripping unit.

Similar nitrogen recovery rates referred to the influent to the recovery unit, ranging between 80 and 97%, were also observed in six other studies with similar operating conditions at a pH range between 8 and 11 and temperatures between 50 and 80 °C (Sengupta *et al.* 2015). Corresponding to our study, the higher the temperature and the pH were, the more the recovery rates increased.

3.2. Characterization of renewable fertilizer qualities

3.2.1. Struvite as high-quality fertilizer

The main components of struvite were TP, Mg and TN with their medians at 12.5, 11 and 10 wt%, respectively (Figure 5). The content of TP and Mg corresponded to their expected values of 13 and 10 wt%, respectively. However, for TN the expected content was 6 wt%, which is far below the measured content of 10 wt%. The authors assume that during sample processing for the TN analysis, the water of hydration volatilized and dehydrated dittmarite formed as described by Saerens (2020). Hence, the TN increased relatively. The TN content of dehydrated dittmarite is 10 wt% which corresponds to the range of the measured values.

The values in blue color in Figure 5 indicate the thresholds for which the components must be declared in the fertilizer's official composition. For TP, Mg and TN, the thresholds of mandatory labeling of those components of 4.4, 0.18 and 1.5 wt% (DüMV 2012) were exceeded and thus, the three components have to be declared. The contents of Ca, K, Na, Fe, S, Al, Mn, B and Co were below their thresholds for a mandatory declaration as well as for Zn, Cr, Cu, As, Ni, Pb, Hg and Cd. Unfortunately, the limit of quantification for Tl was as high as its threshold for pollutants and even higher than its threshold for declaration. Because its content was always below its limit of quantification, it was still below the pollutant threshold. For As, Ni, Pb, Hg and Cd, their contents were far below their thresholds for pollutants (DüMV 2012). Hence, the composition of struvite showed a high quality as fertilizer. For struvite samples resulting from a shorter time period with only four samples, Remy *et al.* (2023) conducted a chemical risk assessment. Besides the heavy metals, for benzo(a)pyrene and PCDD/F + dl-PCB, the risks for the soil system and groundwater ecosystem through the application of struvite as fertilizer were negligible and acceptable confirming the high quality of the fertilizer.

The crystal sizes of struvite ranged between 0.5 and 2 mm being greater than the required sizes by the German Fertilizer Regulation (DüMV 2012) with 0.63 and 0.16 mm for 98 and 90% of the struvite crystals, respectively. Hence, to promote better such technologies in Germany, a wider range of crystal sizes should be included in the regulation. Since 2021, struvite recovered from sewage sludge and wastewater has been included in the European Fertilizing Products Regulation (EU 2021/2086). For Zn, Cu, As, Ni, Pb, Hg, and Cd the thresholds are quite similar to those in the German Fertilizer Regulation (DüMV 2012) and their actual content was far below their thresholds. However, the Fertilizing Products Regulation requires

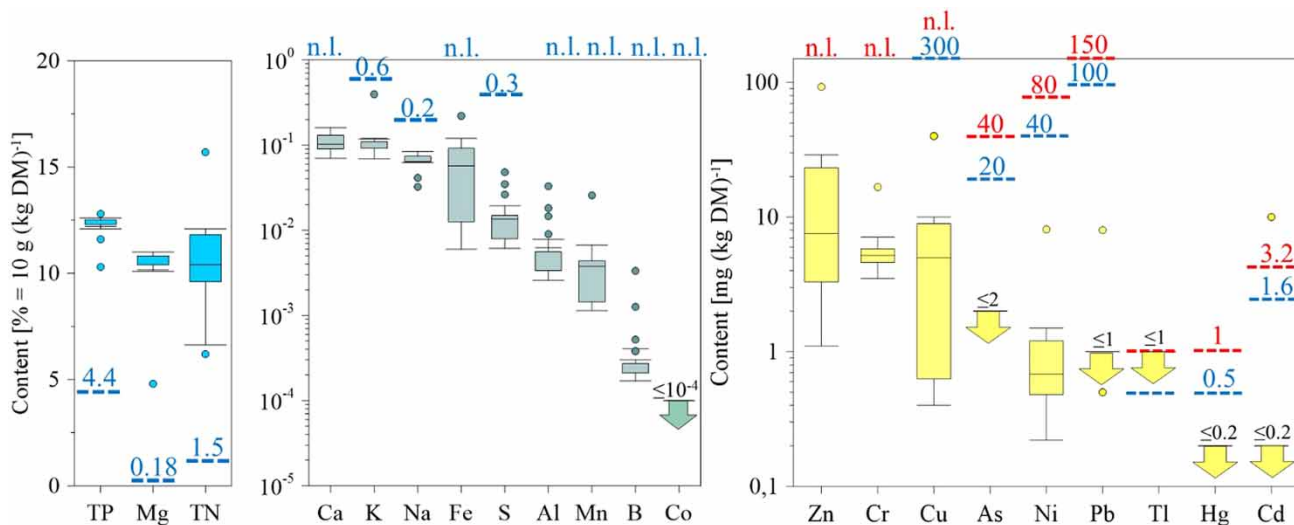


Figure 5 | Characterization of struvite composition in comparison with thresholds for the mandatory declaration of certain compounds (values in blue) and thresholds for pollutants (in red; second value above the threshold for mandatory declaration): $n = 3$ samples for Hg; $n = 12$ for TP, Mg, TN, Ca, K, Na; $n = 13$ for Tl, $n = 24$ for S; $n = 25$ for Fe, $n = 27$ for the rest.

compliance with additional parameters such as hexavalent chromium, biuret and perchlorate, which were not determined in our study. Because the struvite will be applied in Germany, the German Fertilizer Regulation was prioritized.

3.2.2. Ammonium sulfate solution as high-quality fertilizer

The chemical composition of the ammonium sulfate solution showed nitrogen contents of 10 and 9 wt% for TKN and $\text{NH}_4\text{-N}$ (Figure 6) as expected indicating a well concentrated solution and complying with the German Fertilizer Regulation (DüMV 2012) requiring a content of 5 wt% and higher. The contents of Ca, K, Cu, Zn, Mg, TP, Pb, Cr, Ni and Cd were far below their thresholds for an official declaration of their content in the fertilizer's composition as well as for the content of the pollutants (DüMV 2012). However, for half of the samples, a declaration of their Hg content was required and even for 25% of the samples the Hg content exceeded the legal threshold indicating that the pollution with Hg was too high. However, the high Hg levels in the ammonium sulfate solution originate most likely from contaminated sulfuric acid, as even worst-case estimates for sludge liquor Hg content would not result in such high concentrations in the product.

Thus, it is highly recommended to check the quality of process chemicals such as sulfuric acid, before it is used in such a process. In conclusion, besides the critical Hg concentration in the ammonium sulfate solution, the ammonium sulfate solution is a high-quality product. Until now, the Fertilizing Products Regulation (EU 2019/1009) does not explicitly consider ammonium sulfate solution recovered from sewage sludge or wastewater. Remy *et al.* (2023) conducted a chemical risk assessment for the application of the ammonium sulfate solution from Brunswick as fertilizer and its impact on the soil system and groundwater ecosystem referring to a shorter time period (nine samples) than in this study. The assessment showed only negligible and acceptable risks and confirmed the high quality of the fertilizer.

3.3. Carbon footprint

The CO_2e -footprint of the CE system was $16.4 \text{ kg CO}_2\text{e (PE a)}^{-1}$ and similar to the baseline with $16.6 \text{ kg CO}_2\text{e (PE a)}^{-1}$. The main contributor was in both scenarios the electricity demand for wastewater treatment followed by direct emissions (N_2O). It should be noted that the electricity demand for Germany in Ecoinvent v3.6 is relatively high with $617 \text{ g CO}_2\text{e (kWh}_{\text{el}})^{-1}$. The gross emissions were approximately $40 \text{ kg CO}_2\text{e (PE a)}^{-1}$. The credits originated mainly from gained electricity in the CHP through biogas utilization. The CE system improved the overall electricity balance due to the higher biogas yield and reduced efforts in the secondary treatment due to the removal of ammonium from the return load. Thereby, also direct emissions (N_2O) were expected to be reduced (N_2O was calculated by using a linear factor of 0.6% from N in return load). The CE system also gave CO_2e credits for the struvite and ammonium sulfate solution (ammonia was credited as a product of the Haber-Bosch process) by substituting equivalent mineral fertilizer production. Additional impacts came from the chemicals used in the sludge treatment and for nutrient recovery, especially from the additional polymer dosing for pre-dewatering and

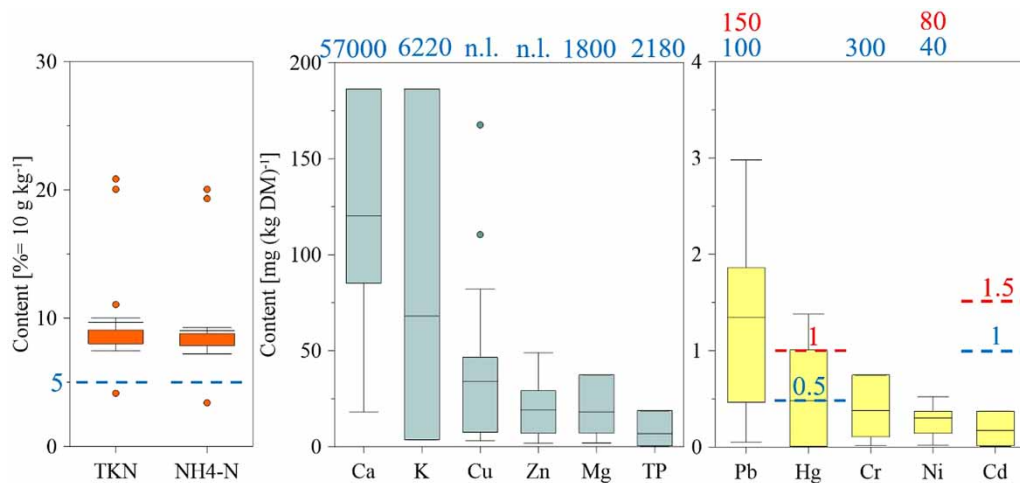


Figure 6 | Characterization of ammonium sulfate solution in comparison with thresholds for the mandatory declaration of certain compounds (values in blue) and thresholds for pollutants (in red; second value above the threshold for mandatory declaration): $n = 18$ samples for TKN; $n = 24$ for $\text{NH}_4\text{-N}$; $n = 23$ for Ca, K, Cu, Zn, Mg, TP, Pb, Cr, Ni, Cd; $n = 20$ for Hg.

NaOH for a pH increase. Furthermore, the higher degradation in the second digestion stage resulted in a lower heating value of the dewatered sludge, decreasing energy credits in the final incineration of the sludge. Further optimization of the CE system might be achieved by utilization of excess heat in summer to operate the ammonia stripper at a higher temperature and hence, to allow for the operation at a lower pH in order to further reduce its NaOH demand.

Overall, the CE system reduced the direct emissions of the system, meaning less N_2O resulted from the secondary treatment due to a lower N return load, and less N_2O resulted from sludge incineration due to a lower sludge quantity after the enhanced digestion. This reduction of direct emissions will be crucial in the future when targeting climate neutrality because the indirect emissions from electricity, heat and chemical production will decline due to a more renewable energy mix in Germany. Therefore, the direct emissions will become more relevant for the net CO_2e -footprint. The reduction of direct emissions and a more targeted application of reactive N species (as mineral fertilizers) can thereby play a crucial role in climate neutrality in wastewater treatment.

Whereby LCA studies looked either at P recovery (Lam *et al.* 2020) or N recovery (Kar *et al.* 2023; Saud *et al.* 2023), this study looks holistically from the WWTP perspective at a combination of both, together with a higher biogas production.

The CE system improved the energy efficiency of the WWTP, due to the higher recovered CH_4 yield. However, holistically the entire sludge disposal's energy recovery was reduced due to the lower heating value and thereby lower heat recovery in sludge incineration. Nonetheless, the DM content after sludge dewatering and the sludge disposal route, and its electric and thermal efficiencies were crucial for the overall energy efficiency of the CE system with TPH.

While the net impact regarding the efforts for phosphate recovery of the CO_2e -footprint is lower compared to that of nitrogen recovery and the TPH (see Figure 7), the results generally fit with the meta-study conducted by Lam *et al.* (2020). However, the credits associated with nitrogen recovery and the TPH due to reduced efforts for nitrogen removal in the secondary treatment and increased biogas production are severely higher than for phosphate recovery. A direct comparison of carbon footprints is not possible, due to different functionalities chosen by Saud *et al.* (2023). Nevertheless, NaOH is also mentioned by Saud *et al.* (2023) as one main contributor to the CO_2e -footprint. This is also confirmed within this study, as the main contributor in terms of chemicals is NaOH to increase the pH for ammonia stripping. Kar *et al.* (2023) compare several stripping plants with a similar heat demand as in our study. Neither Saud *et al.* (2023) nor Kar *et al.* (2023) reflect the

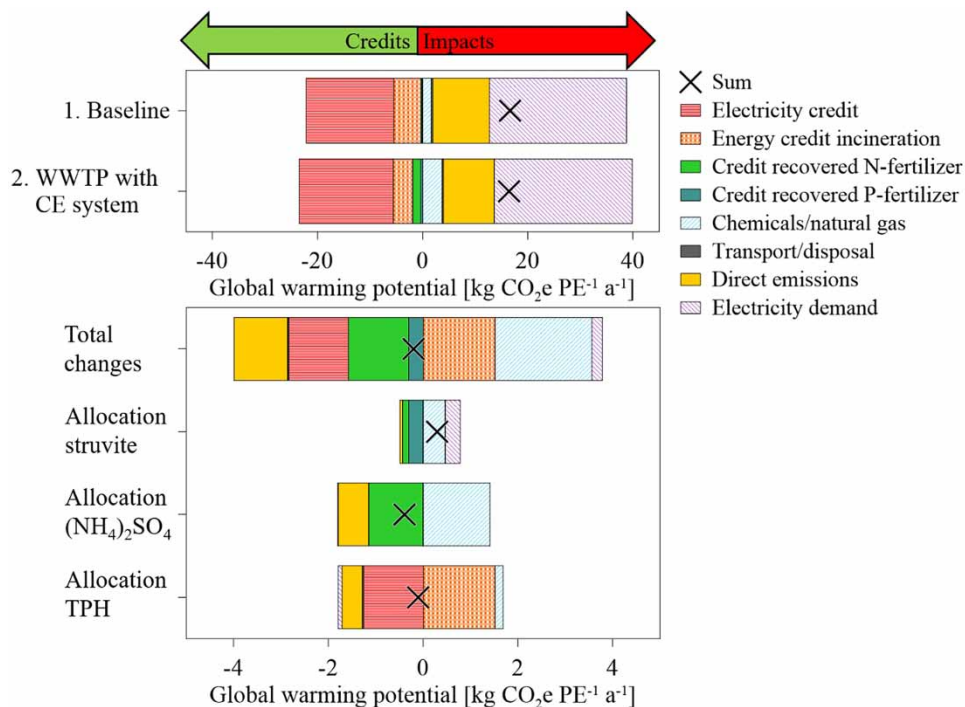


Figure 7 | Global warming potential for the (1) baseline (WWTP without CE) and (2) WWTP with CE system and the changes, when comparing both scenarios. In addition, the changes are allocated to struvite production, ammonium sulfate $(NH_4)_2SO_4$ production and the TPH.

positive impact of reduction of the nitrogen removal in return load on N_2O emissions in secondary treatment, which is a main advantage of this process as shown in our study.

Further optimization measures should be investigated to valorize available excess heat in summer, especially when the valorization of the digestate in agriculture via fertigation will end in 2028 and higher nitrogen return loads will be expected to achieve even higher process temperatures ($>65\text{ }^\circ\text{C}$) in the stripper and thereby to further reduce NaOH consumption.

The carbon footprint for the entire system decreased slightly towards carbon neutrality thanks to the new system because more biogas and renewable fertilizers were produced and nitrous oxide emissions were reduced. Using a green energy mix only in the future, the latter will be crucial to reach carbon neutrality.

4. CONCLUSIONS

The combination of a DLD system with nutrient recovery units offers several synergies and only one small drawback:

- The increase in the methane production rate of 18% showed a higher solubility and biodegradability of the hydrolyzed sludge. This suggests also a higher release of ammonium and phosphate from the hydrolyzed sludge, which will be a benefit for the subsequent nutrient recovery units.
- The increased dewaterability leads to a reduction in costs for the dewatered digestate, because its volume decreases with a higher density.
- Even though the results suggested the formation of refractory COD, which is a drawback of thermal hydrolysis, the increase was so low with 4 mg L^{-1} , that the legal requirements for the WWTP effluents were still met. In summary, the innovative combination of the technologies contributed to a better effluent quality in terms of nitrogen and phosphorus of the WWTP and hence, the system is very well suited to treat higher nutrient loads (factor 1.3) than the WWTP was originally designed for.
- The NuReSys system for struvite production delivered reliable recovery rates of 80% and greater as long as the molar ratio of Mg:P for the MgCl_2 dosing was maintained at 0.8 and higher. The struvite process is crucial to be in operation in order to prevent the subsequent ammonia stripping process from failure through the formation of undesired precipitants and scaling processes for example in the heat exchangers, which was observed at the beginning of the operational phase.
- High ammonium recovery rates were reached also at lower pH values around 8.5, when the process temperature was maintained at $65\text{ }^\circ\text{C}$. This is especially rewarding for systems, where excess heat is available and can be used to maintain the process temperature so that NaOH can be saved.
- The composition of the secondary fertilizers showed, a high quality of the products. Hence, the sludge liquor resulting from municipal wastewater treatment and sludge management as a source for the products contributed to its high quality and was suitable for secondary fertilizer production. However, when using additional chemicals for producing a fertilizer such as sulfuric acid, its quality needs to be controlled to make sure that it is free of pollutants.

The carbon footprint showed that the overall energy balance of the TPH was positive, however:

- This depends highly on the assumptions for the DM content after sludge dewatering and in the sludge valorization route.
- The impact on the energy balance for the WWTP is positive assuming sludge is thermally utilized at an external facility.
- In terms of phosphorus recovery, the impact on the carbon footprint is almost negligible.
- NaOH consumption and heat management in the stripping plant are crucial factors influencing the carbon footprint.

For future optimization available excess heat in summer should be utilized to achieve process temperatures above $65\text{ }^\circ\text{C}$ in the stripper in order to reduce NaOH consumption.

The new CE system reduced the carbon footprint slightly. In the future, a greener electricity mix will highly contribute towards the carbon neutrality of the system.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its supplementary material.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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