Innovative Treatment of Municipal Sewage Sludge Using Hydrothermal Carbonization and Nutrient Recovery Technologies

Utilization Potential in the DACH Region and a Short Evaluation of the Environmental Impacts

Marion Andritz(✉)

Chair of Process Technology and Industrial Environmental Protection Montanuniversitaet Leoben, Franz-Josef-Straße 18, 8700 Leoben, Austria
marion.andritz@unileoben.ac.at

Abstract. Phosphorus (P) is an essential and at the same time critical resource, as there are barely any available reserves in the European Union, which means that there is a dependence on imports from non-EU countries. In these countries, there are critical working and environmental conditions for which social responsibility must be taken. To address these challenges, municipal sewage sludge has to be utilized since it has a huge potential to produce sustainable P- fertilizers. Hydrothermal Carbonization (HTC) is an energy-improved treatment of sewage sludge and enhances the dewatering properties. The remaining process water contains a significant nutrient content, especially nitrogen (N) and phosphorous (P). For a future project, the recovery potential of P was investigated for Germany, Austria and Switzerland (DACH region). The forecasted P recovery potential in the DACH region until 2030 covers 25% of the annual P demand and the environmental impact due to the conventional industrial production of P fertilizer could be reduced dramatically (up to 63 t of CO_2 emissions can be saved per year). To identify the sustainable development goals (SDGs) which are primarily addressed by the project, the following question was formulated: What effects does the technology have in terms of sustainability (social, ecological, economic)? The investigation showed that SDGs 8, 11, 12, and 13 are mainly focused. The investigations serve as a basis for a future project, which is shortly introduced in this work, where the nutrients, as well as heavy metals, will be recovered by an innovative combination of three novel processes.

Keywords: Wastewater treatment · Phosphorus recovery · Ion exchange · Zeolite · Sustainable consumption and production · Fertilizer

1 Introduction

In recent years, the utilization of sewage sludge (SS) has been heavily regulated, which currently confronts many wastewater treatment plants in Germany, Austria, and Switzerland, and thus cities and municipalities with rising wastewater disposal costs. The majority of sewage sludge is incinerated, which shows that the focus is on energy yield or

J. A. Benítez-Andrades et al. (eds.), *Global Challenges for a Sustainable Society*,
Springer Proceedings in Earth and Environmental Sciences, https://doi.org/10.1007/978-3-031-25840-4_19

secure disposal, but not on the waste management principles of closing the loop and creating safe sinks. Meanwhile, the Austrian Federal Environmental Agency's Waste Management Plan, for example, calls for a […*need for action in the area of sustainable phosphorus recycling from sewage sludge and animal meal…]* and describes new technologies to condition sewage sludge and recover the phosphorus it contains [1].

Phosphorus is an irreplaceable element that is needed for plant growth and therefore plays an important role in fertilizer production. The reasons for phosphorus recycling are manifold, sometimes the declining quality of phosphate reserves plays a role, as they are increasingly contaminated by heavy metals and represent an environmental problem. There is also a case for securing supply independence since Europe itself has no significant deposits and up to 90% of the economically exploitable phosphate reserves are located in Morocco, China, the USA, Algeria, Jordan, Russia, and South Africa [2]. Since the European Union classified P as a critical raw material, especially in terms of economic importance, innovative and new technologies for P recovery must be implemented [3]. As a result, conventional sewage sludge treatment, for example, co-incineration of sewage sludge in waste incineration plants, cement, or coal-fired power plants will no longer be possible [4].

In addition, in the countries of primary P fertilizer production, there are inhumane working conditions and critical environmental impacts, such as the disposal problem of phosphate gypsum contaminated with heavy metals and radioactivity. To meet the United Nations Sustainable Development Goals (SDGs), municipal sewage sludge must be utilized since it has a huge potential to produce sustainable P fertilizers [5, 6]

1.1 Current Situation in Terms of Technologies for Sewage Sludge Treatment and Phosphorous Recovery

To meet recent requirements in terms of phosphorus recovery, technologies for sewage sludge conditioning and nutrient recovery are currently being promoted in industry and research, but they differ greatly in their technical concept and recovery rate, as well as in their technical readiness level. The only concrete technical measure mentioned is the mono-incineration of sewage sludge with subsequent P recovery from the incineration ash. This technology also brings aspects such as high consumption of chemicals or long transportation distances, as well as the monopoly position of waste management companies with centralized treatment plants. The problem becomes even more obvious that there is a gap between decentralized sewage sludge production and centralized sewage sludge utilization that needs to be closed [7].

The BAT (best available technology) for waste incineration plants also does not mention alternatives to P recovery from ash, while the report of the Federal Ministry (BMLFUW) proposes decentralized, on-site treatment and recovery of P, but also mentions thermal recovery in a central sewage sludge mono-incineration as a future option [8].

An alternative is the conditioning of sewage sludge using pyrolysis or hydrothermal processes. Both processes are characterized by the fact that they are coupled to wastewater treatment plants and sewage sludge with a high water content ($>$ 70 vol.%) can be treated. Accordingly, conditioning and disposal costs are reduced, and the resulting process gas is lower in volume compared to conventional incineration.

However, pyrolysis entails significant disadvantages, such as the required tightness, inert gas atmosphere, operational safety (T > 600 °C), and by-products, such as coke or pyrolysis oil, whose use is severely limited due to their composition [9]. Much more efficient and operationally reliable process management is offered by the treatment of sewage sludge with HTC processes (hydrothermal carbonization), which is one of the future technologies for energetically optimized treatment [10].

1.2 Hydrothermal Carbonization

Hydrothermal Carbonization (HTC) is an energy-improved alternative for the treatment of sewage sludge and enhancing the dewatering properties. This novel approach treats sewage sludge with a water content of up to 75% at approx. 220 °C and 24 bar, resulting in a thermochemical conversion. Here, ,nature's natural carbonization is recreated in a technical process. Dehydration and carboxylation processes produce a brown-to-black solid with the calorific value of peat or lignite and a lower oxygen and hydrogen content. The process significantly improves the mechanical dewatering properties and at the same time sterile the sludge. As a result, sludge volumes are significantly reduced compared to other mechanical dewatering methods [11, 12].

The filtrate after mechanical dewatering is referred to as HTC process water and has a high proportion of nitrogen and phosphorus, the residue is referred to as HTC char/coal and is characterized by a high amount of carbon. The process can be controlled by process conditions, like pressure, temperature, pH, heating rate, or time, as well as the additives in such a way that the division of the nutrients into solid and aqueous products can be adjusted. Compared to conventional drying and dewatering processes, the HTC process has advantages due to the better dewatering properties of the sewage sludge after mechanical dewatering and thus a lower total energy demand. In addition, there is a positive CO_2 balance and cost efficiency (on-site concept, reduction of transport, storage and disposal costs) [13].

Thus, the remaining process water contains a high concentration of organic and inorganic compounds (COD content up to 10.000 mg/L)1, but also a significant nutrient content, nitrogen (N), and especially phosphorous (P). Therefore, the challenge is to utilize the process water for nutrient recovery and both, isolate the critical components to meet the requirements for the discharge availability of the remaining process water [14].

2 Objectives

Within this work, the phosphorous potential for recovering from municipal sewage sludge in Germany, Austria and Switzerland (DACH region) has to be investigated and an assessment to show the impact on the environment has to be carried out. The last step is to identify those key SDGs to present them as relevant and considerable. Based[1] on the previous tasks, a future project should be outlined, which addresses the treatment

[1] Analyses of real HTC process water from a plant in Switzerland, carried out by the author Marion Andritz (01/2021).

of municipal sewage sludge and the recovery of nutrients from the process water after HTC treatment.

The project has not started yet, and it is also not part of this paper, however, it is shortly introduced by the scheme in Fig. 1 to understand the considerations in Chapter 3. The scheme demonstrates a possible process combination, where phosphorous as phosphate, nitrogen as ammonia, as well as heavy metals, are to be recovered after the hydrothermal treatment of sewage sludge. Therefore, the ion-exchange loop stripping process (ILS process) with a natural zeolite [15] and the FerroDECONT process (a redox process with zero-valent iron) were combined [16].

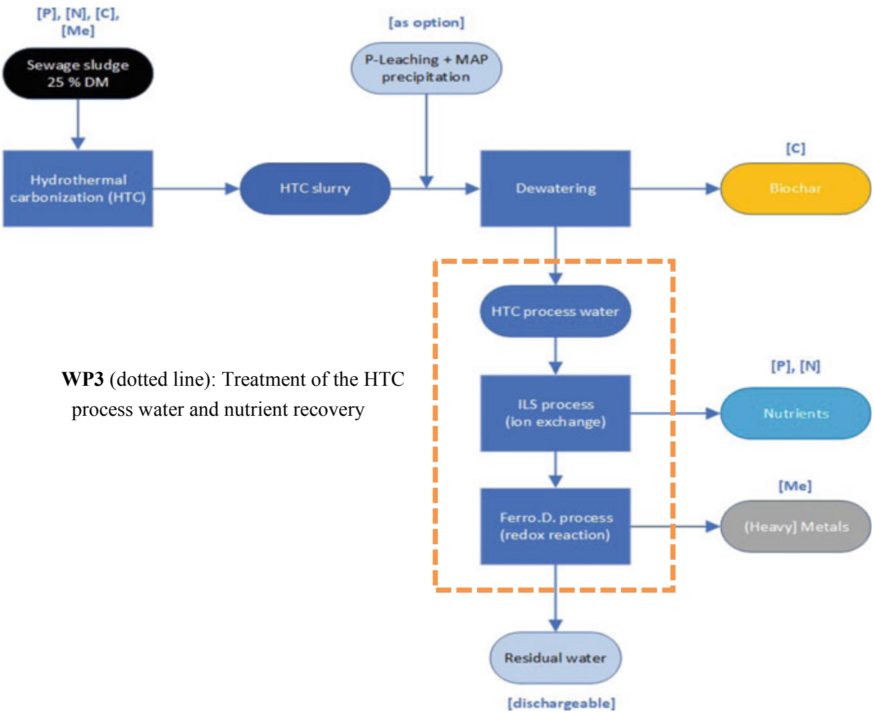

Fig. 1 Scheme of the process: HTC process water treatment with the ion-exchange-loop- stripping process for nutrient recovery combined with the ferroDECONT process for metal isolation

The products are a nutrient-rich fertilizer solution and a low pollutant but carbon- and energy-rich biochar which can be utilized for material and energy purposes. The scope of the project is to develop an overall concept that serves as a decision-making basis for the technical implementation of these processes in an existing wastewater treatment plant in Austria. One character of the project is a life cycle analyses of the technology and the task to consider the principles of circular economy which is defined in a separate work package (WP). In total, the project consists of 5 WPs whereas the WP3 focuses on the treatment of the HTC process water and nutrient and metal recovery and the WP4 considers the sustainable perspectives, which this paper is part of it.

3 Methods

3.1 Assessment of the Phosphorous Recovery Potential for the DACH Region

To assess the benefits and impact of phosphorus recovery strategies, the potential for Germany, Austria and Switzerland (DACH region) was evaluated by the author. The forecast runs until 2030, as it is assumed that by then the strategies will be legally anchored and the technologies regarding the recovery rates will be mature. The reason for choosing these countries is the availability of meaningful data basis on previous research.

Sewage sludge volume: In the first step, the annual amount of sewage sludge was determined in terms of dry matter (DM) and population equivalent (PE). However, to be able to make reliable data for a current potential estimate or an estimate up to the year 2030, some assumptions were made, which are explained in the following.

In Germany, the total sewage sludge volume in 2010 was around 1.89 million [t] dry matter. In 2018, the amount of sewage sludge produced was approx. 1.74 million [t] dry matter and was thus roughly the same as in previous years. According to statistics on the development of municipal sewage sludge production in Germany from 2010 – 2019, the amount of municipal sewage sludge has not fluctuated significantly in Germany over the last 10 years [17].

The volume of municipal sewage sludge in Austria has remained in the same order of magnitude in recent years and has always been in the range of 235,000 to 238,000 [t] dry substance [18].

Compared to Austria and Germany, there are no current central statistics in Switzerland on the development of sewage sludge production or the types of treatment. These data are documented in the individual cantons, and in some cases, no distinction is made between municipal and industrial sewage sludge, or between sludge from other sources. To be able to make statements about the volume and utilization of sewage sludge, the data from 2017 were used in current publications. According to [19] the sewage sludge volume in 2017 was around 178,000 [t] dry matter.

To collect an estimate of the potential for the DACH region, the period 2017–2018 was therefore chosen as the reference period, based on extensive data for Switzerland. **Usable amount of sewage sludge for P recovery**: For the actual usable amount of sewage sludge that can be considered for future recovery strategies, the previous recovery sectors in the DACH region were surveyed within the scope of research, whereby it was shown in summary that in all 3 countries about 80% of the sewage sludge quantities can be considered for P recovery. This includes those quantities that are already thermally utilized and will be treated in the future in a mono-incineration with P recovery from the ash. Another part is those quantities that are currently still landfilled or agriculturally utilized and, in both cases, will be prohibited and strictly regulated by 2030 and thus must also be subjected to treatment. The remaining 20% was accounted for by other disposal routes, such as co-incineration in cement plants, whereby it is assumed it will remain unchanged [20].

The phosphorous content in municipal sewage sludge: According to [21], sewage sludge contains between 20.0 and 25.0 g P/kg DM. It should be mentioned that the calculations are based on elemental phosphorus. For further estimation, the mean value (22.5 g P/kg DM) was used. Multiplied by the annual utilizable amount of sewage sludge in Germany, Austria and Switzerland, the theoretical amount of phosphorus that could be theoretically recovered is obtained.

Recovery rates from sewage sludge: According to a report by the DECHEMA [2], there are currently about 70 processes for P recycling from sewage sludge and sewage sludge ash, which vary greatly in terms of efficiency or recovery rate. The final report of the "StraPhos" study [4] showed that the rate depends not only on the respective technology status but also on the principle of recovery (from the ash after mono- incineration or wet-chemical), as well as on the input quantities. Especially whether the recovery takes place decentral at the wastewater treatment plant with lower input quantities ($< 50{,}000$ PE) or centrally in a mono-incineration plant with input quantities ($> 100{,}000$ PE). Therefore, the report shows that the efficiency for decentralized concepts is between 45 and 70% and for centralized technologies between 60 and 85%. For further calculations, the respective average values for the former with 58% (decentralized, wet-chemically) and the latter with 75% (centralized, thermochemically) are used. In further surveys, the share of wastewater treatment plants that can be considered for a decentralized or a centralized recovery strategy was estimated. According to this, a classification of the size classes in Austria and the resulting sewage sludge quantities shows that 1/3 can be used for decentralized recovery strategies and 2/3 for centralized technologies. This approach was approximated for the entire DACH region [21].

Demand for phosphorous for fertilizer production: The demand for phosphorus merely for agricultural use as fertilizer is based on surveys from AMA Austria [22] and was assumed to remain constant in 2030 according to the annual statistics. Not included in the data was the demand for additives in animal feed or chemicals [23].

3.2 Evaluation of the Environmental Impacts

For the environmental impacts, the overburden produced during phosphate mining, the phosphorus gypsum produced, and the possible CO_2 savings are surveyed. Existing life cycle assessments show that between 7 and 10 kg of phosphate rock is mined for every kg of pure phosphorous [kg rock/kg P]. During the production of phosphorus fertilizer, between 4 and 6 kg of phosphate gypsum is produced per kg of phosphorus, which is contaminated with heavy metals and radioactive substances and is currently landfilled. The CO_2 emission results from the input of fossil energy into the operation. In addition to fossil energy sources, this also includes process energy for the production and distribution of mineral fertilizers, pesticides, electricity, water, and external services and varies between 0.5 and 2.4 kg CO_2/kg P [24].

The yearly amount of conventional phosphorous that could be replaced is then multiplied by the previous ranges to estimate the environmental effects.

3.3 Relation to the Sustainable Development Goals (SDGs)

The relationship between phosphorus recovery from municipal sewage sludge and the SDGs based on the definitions by the United Nations [25] was considered and qualitatively formulated by the author. For the first approach, a stakeholder analysis was conducted to assess the benefits and impacts.

It was assumed that at a wastewater treatment plant with < 50.000 PE the sewage sludge treatment is carried out by hydrothermal carbonization (described in 1.2) followed by a wet-chemical phosphorus recovery process (for example an ion exchange process). This means that the sewage sludge is processed directly on-site. Therefore, there are no additional disposal or transport costs to other treatment companies. The phosphorus recovered is in a condition (unspecified) that it can be used as a feedstock for further fertilizer production (not on-site). The technology system is compact (container size) and can be easily installed on the existing wastewater plant. Accordingly, the plant includes conventional (existing) wastewater treatment processes, innovative sewage sludge conditioning with HTC, and a process for nutrient recovery, where the latter can be considered as an additional sales product to the fertilizer sector.

The next step is to identify the key SDGs which are relevant and considerable. In this way, it can be shown which SDGs the technology has the greatest influence. A document from [26], based on [27] was used as a guideline to conduct a simple but robust analysis. Therefore, three questions (see Table 1) were developed by the author to identify the key SDGs. The answers and ideas were then collected and compared with the contents of the SDGs. The following questions were defined:

Table 1 Questions to answer for identifying the key SDGs (defined by the author)

Questions
What effects does the technology have in terms of sustainability (social, ecological, economic)?
Where are points of contact with adjacent actors (e.g. municipalities)?
Where and in which way is an external impact generated (regionally, globally)?

4 Results

4.1 Potential for DACH Region

In 2030, the total volume of sewage sludge will be up to 2.153 [t] per year, where 1.722 [t] can be utilized for phosphorous recovery and 39 [t] per year will be recovered, based on the assumption that the recovery technologies are on technology readiness level higher than 7 [4]. **Table 2** summarizes the results for the DACH region based on the assumptions in 3.1. The potential survey indicates that up to 25 wt% of the annual phosphorus demand to produce fertilizers in the DACH region can be replaced.

Table 2 P-recovery potential [kt/a] from sewage sludge (SS) for the DACH region until 2030

Parameter	Value [t/a]
Volume total of SS in DM	2153
The volume of SS in DM used for P recovery (80%)	1722
The potential amount of P (elementary) in SS (22,5 g P/kg DM SS)	39
Recovery potential (averaged rate, depending on technology and plant size)	26
The demand for P for fertilizers in agriculture (excl***animal feed, chemicals)***	105

4.2 Environmental Benefits

The environmental effects due to conventional phosphorus being replaced by recovering are shown in Table 3.

Table 3 Environmental benefits by replacing conventional P fertilizer production with recovered P in the DACH region until 2030

Parameter of environmental impact	Unit	Statistical values (range)	Total reduction/benefit (range) [t/a]
Reduction of overburden	[kg rock/kg P]	7–10	185–264
Reduction of harmful gypsum to\be landfilled	[kg gypsum/kg P]	4–10	106–160
Reduction of CO_2 emissions	[kg CO_2/kg P]	0.5–2.4	13.2–63

Up to 264 [t] of overburden produced during phosphate mining can be reduced and 160 [t] of the harmful phosphorus gypsum can be reduced. The possible CO_2 savings are between 13.2 and 63 [t] per year and vary greatly since the system boundaries are not uniformly set.

4.3 Relation to the Sustainable Development Goals (SDGs)

Based on the previous analysis the technologies of decentralized hydrothermal carbonization of sewage sludge with subsequent recovery of phosphorus primarily address the following goals:

SDG 6. Clean water and sanitation: Recovery processes contribute to wastewater disposal by relieving the burden on sewage sludge recycling and creating an alternative use for valuable sewage sludge components, like nutrients. As a result, wastewater disposal become more efficient and sustainable. Further, since the sewage sludge treatment and the nutrient recovery is located on-site, it can also be implemented worldwide, especially in less developed regions.

SDG 8. Decent work and economic growth: On the part of the EU, economic dependencies are reduced since sewage sludge is available in large quantities and all year round. Additionally, the national added value and economic performance are increased through the creation of new technologies and thus jobs. For companies in the field of mechanical and plant engineering, the implementation of the technology also leads to the expansion of the portfolio through innovative technologies and the promotion of expertise. Consequently, jobs are also created in this area.

The majority of phosphate mining takes place in North Africa and the Middle East, with inhumane working conditions. Insufficient protective equipment leads to respiratory, and heart problems, and cancer. Phosphorus recovery can reduce phosphate degradation and in addition, new jobs will also be created in those regions if phosphorus recovery technology is expanded globally. This guarantees to take social responsibility for less developed countries.

SDG 11. Sustainable Cities and Communities: Growing cities will have to deal with increasing amounts of wastewater and sewage sludge. The treatment of sewage sludge with HTC processes enables the reduction of sewage sludge volumes and the recycling of previously unused side streams. Wastewater treatment plants become deliverers of sustainable nutrients and renewable energy by using biochar energetically. and reducing transport distances. Efficient and economic sewage sludge utilization is of high social relevance, as it leads to reduced odor irritation, reduced transport frequency, and noise pollution. In addition, the technology makes a significant contribution to reducing wastewater charges and municipal disposal costs. This leads to the minimization of financial burdens (sewer fees) for all affected households.

SDG 12. Sustainable consumption and production: The provision of nutrients and energy from residues (sewage sludge) increases resource and energy efficiency and reduces the impact of primary production. This includes the fact that the process water produced during conventional phosphate mining is discharged into the surrounding waters, and large quantities of Phosphate gypsum are landfilled as a partly radioactive by-product. The process heat generated by the HTC process can be recovered, and no chemicals are required for the process itself. The use of sewage sludge as a source of renewable energy (fuel or biogas) and nutrients leads to a simultaneous reduction of greenhouse gas emissions. This means that an environmentally damaging effect is avoided. The space requirements for both processes (HTC and nutrient recovery process) are significantly lower than for central mono- incineration plants. Since these processes are used in a decentralized manner, the surrounding region also benefits from them, as the recovered nutrients can be further processed for fertilization in local agriculture.

SDG 13: Climate action: By implementing the technology, it can be demonstrated that a contribution to climate protection can also be made in an existing wastewater treatment plant. In addition, education, and openness for sustainability in the region are promoted by such technologies (lighthouse project).

In a future study, which is not covered in this paper, a quantitative and measurable monitoring system will be developed to evaluate the achievement of the considered SDG goals.

5 Conclusions

In this paper, an estimation of the annual amount of phosphorous (P) which can be theoretically recovered, and the impact of the environmental aspects were carried out. Further, the relationship between the SDGs and the phosphorus recovery from municipal sewage sludge was evaluated and showed that SDGs 8, 11, 12, and 13 are the key goals. In a future study, a quantitative and measurable monitoring method has to be developed to evaluate the achievement of the considered SDG goals. The connection to the SDGs and following those goals thus serves as an effective orientation to ensure a positive contribution to the environment and humans.

The forecasted P recovery potential in the DACH region until 2030 replaces not more than 25% of the annual demand for elementary phosphorous [t/a], but the environmental impact could be reduced dramatically. Up to 264 [t] of overburden produced during phosphate mining can be reduced and 160 [t] of the harmful phosphorus gypsum can be avoided. The possible CO_2 savings are between 13.2 and 63 per year, which vary greatly and depend on the set system boundaries of the regarded process. However, the results show positive environmental effects, which are pronounced for overburden and landfilling. By avoiding overburden and phosphor gypsum, huge areas of green spaces, landscapes, and forests, thus habitat, can be protected. In addition, the input into the groundwater is reduced by the substances contained in the gypsum. Thus, today the potential is limited to the current state of the art of technologies which are mostly on a demonstration or pilot plant scale [2].

The recovery technologies must guarantee high efficiency and need further and intensive research activities, especially including wastewater treatment plants and municipalities to guarantee an overall concept that can be realized. A significant aspect is, that wastewater and sewage sludge are not only regarded as a “sink” but as a “source” of nutrients and follow the principle of the circular economy. Based on the above potential surveys and sustainability assessment, a future project will be outlined, which addresses the treatment of municipal sewage sludge and the recovery of nutrients from the process water. Therefore, in the next step, the first experiments will be carried out and the nutrient and metal recovery process will be investigated.

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