detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

HYDROTHERMAL CARBONIZATION OF SEWAGE SLUDGE – AN EFFECTIVE APPROACH TO TREAT AND MANAGE SEWAGE SLUDGE IN RURAL AREAS OF GERMANY?

Tommy Ender [1,*], Vicky Shettigondahalli Ekanthalu [1] and Michael Nelles [1,2]

[1] *Department of Waste and Resource Management, Faculty of Agricultural and Environmental Sciences, University of Rostock, D-18059 Rostock, Germany*
[2] *Deutsches Biomasseforschungszentrum gGmbH (DBFZ), D-04347 Leipzig, Germany*

Article Info:
Received:
2 February 2023
Revised:
27 July 2023
Accepted:
4 August 2023
Available online:
30 September 2023

Keywords:
Hydrothermal carbonization
Nutrient recovery
Phosphorus recovery
Sewage sludge utilization
Rural areas

ABSTRACT
As the result of new regulation from the German Sewage Sludge Ordinance (AbfKlärV 2017) and the future obligation to recover phosphorus, thermal treatment (mono-incineration) has become increasingly popular, whereas land-based utilization has decreased. Germany has produced 1.71 million metric tons (dry matter) of sewage sludge in the year 2021. Sewage sludge contains important nutrients such as phosphorus but also heavy metals and organic pollutants making the direct utilization of sewage sludge in agriculture controversial. Rural areas in particular have benefited from land-based sewage sludge utilization however the future ban on direct land-based utilization is forcing them to find alternative solutions for sewage sludge treatment and management. Hydrothermal carbonization (HTC) has developed considerably over the last 15 years and offers a viable alternative for the utilization of municipal and industrial organic waste such as sewage sludge. The process takes place in an aqueous environment without the need for pre-drying sewage sludge and thereby facilitating direct processing. HTC is especially suitable in combination with the recovery of nutrients like phosphorus. Technologies to recover this essential resource are important because phosphorus is an element that cannot be substituted and is therefore essential. HTC could make a significant contribution to sewage sludge management in combination with phosphorus recovery. However, the technology has yet to establish itself as a sewage sludge valorization process (2023) and is not yet a recognized state-of-the-art. Nevertheless, the HTC technology could gain greater relevance in the future, especially as an alternative valorization pathway for sewage sludge in rural areas of Germany.

1. INTRODUCTION

Sewage sludge is the waste generated from the treatment of wastewater in wastewater treatment plants (WWTP). The sludge is a by-product in addition to the treated wastewater which contains nutrients and has to be utilized (Huezo et al., 2021). Sewage sludge can be divided into primary, secondary and tertiary sludge (Roskosch et al., 2018). Primary sludge is also known as pre-sludge, is produced through sedimentation in primary clarifiers during the mechanical-physical treatment of effluent. Secondary sludge is divided into return sludge and activated sludge. Secondary sludge is separated from treated wastewater in secondary clarifiers. The sludge produced during further wastewater treatment is called tertiary sludge. For instance, phosphate elimination processes used in downstream chemical purification produce this (Roskosch et al., 2018). The term raw sludge refers to non-stabilized or partially stabilized sludge (AbfKlärV, 2017).

The Federal Statistical Office of Germany indicates that 1,717,803 t DM (dry matter) of sewage sludge was generated in the year 2021 (Destatis, 2023). The current trend of sewage sludge utilization is greatly shifting towards thermal utilization and no longer towards land-based utilization. In addition to nutrients, sewage sludge also contains heavy metals and organic pollutants making its direct application for agriculture purposes difficult (Aragón-Briceño et al., 2021). In Germany, sewage sludge disposal options have become progressively limited as a result of advanced environmental policies and the laws that have been adopted over the last decade. According to the German Sewage Sludge Ordinance (AbfKlärV 2017), sewage treatment facilities larger than 100,000 population equivalent (PE) and 50,000 population equiv-

* Corresponding author:
Tommy Ender
email: tommy.ender@uni-rostock.de

https://doi.org/10.31025/2611-4135/2023.18308

alents (PE) will no longer be allowed to recycle sewage sludge for land-based utilization starting in 2029 and 2032, respectively. About 79% of the total amount of sewage sludge generated in 2021 (1,364,890 t DM) in Germany was thermally utilized (including mono- and co-incineration among other thermal disposals) and the amount of sewage sludge managed through land-based use and landscaping is gradually declining. Figure 1 shows the trend in sewage sludge utilization in Germany from 2015-2021 (Destatis, 2023).

The sewage sludge producers are required by the German Sewage Sludge Ordinance to recover phosphorus from the sludge on a mandatory basis. Phosphorus recovery from the ash or carbonaceous residue is done after the mono-incineration of sewage sludge. Alternatively, the co-incineration of sewage sludge can be done only after reducing the phosphorus content to at least 50% or to < 20 g P/kg DM. The sewage sludge that resulted could only be used thermally in co-incineration (e.g., cement plant, power plant, waste incineration) after meeting this requirement (AbfKlärV 2017; Mix-Spagl, 2017). All sewage sludge producers are required to voluntarily inform the relevant authority by 31st December 2023 of their future plans for recovering phosphorus. An obligation to recover phosphorus will come into existence in the year 2029 for all WWTP that treat municipal wastewater (Montag et al., 2022). Another considerable regulation in the context of land-based utilization is the German Fertiliser Ordinance (DüMV 2012). Among the other regulations, this ordinance regulates the placing on the market of fertilizers that are not designated as EG-fertilizers (Roskosch et al., 2018). Furthermore, this regulation classifies sewage sludge as an organic or organic-mineral fertilizer and makes a provision to be used as fertilizer if it meets the specified limits (Roskosch et al., 2018). In addition to the limit values in the Sewage Sludge Ordinance (AbfKlärV 2017), those in the Annex to the German Fertiliser Ordinance (DüMV 2012) also apply to the agricultural-based utilization of sewage sludge. As a result, the possibilities for direct land-based utilization are limited especially in the case of heavy metals.

The typical composition of sewage sludge is shown in Table 1. Table 1 also depicts the limit values of the German Fertilizer Ordinance (DüMV 2012) and German Sewage Sludge Ordinance (AbfKlärV 2017) for direct land-based applications. If sewage sludge does not comply with the qualitative requirements of the German Fertilizer Ordinance and German Sewage Sludge Ordinance, it has to be recycled in other ways (Langenohl, 2015).

Stringent regulations from the German Sewage Sludge Ordinance, (AbfKlärV 2017) and the future obligation of phosphorus recovery has made thermal valorization increasingly popular whereas land-based utilization and landscaping have decreased. Rural areas in particular have benefited from land-based sewage sludge utilization.

As previously stated, land-based application of sewage sludge was the most common method for managing and utilizing sewage sludge in rural areas. Since this recycling method is about to be phased out, there is growing interest in finding alternative ways to recycle sewage sludge and recover phosphorus. In this concern, thermal valorization (mono-incineration) of sewage sludge in combination with phosphorus recovery from ash might be widely accepted. The primary requirement of sludge to be dewatered and dried however makes the incineration process cost-intensive and poses concerns related to the release of toxic air pollutants (NO_x, SO_2, and dioxins) if the incineration process is not managed properly (Shettigondahalli et al., 2022). Small and medium-sized WWTPs and municipalities will need to find alternative methods of sewage sludge management and treatment in the future if they do not have access to such thermal sludge valorization opportunities.

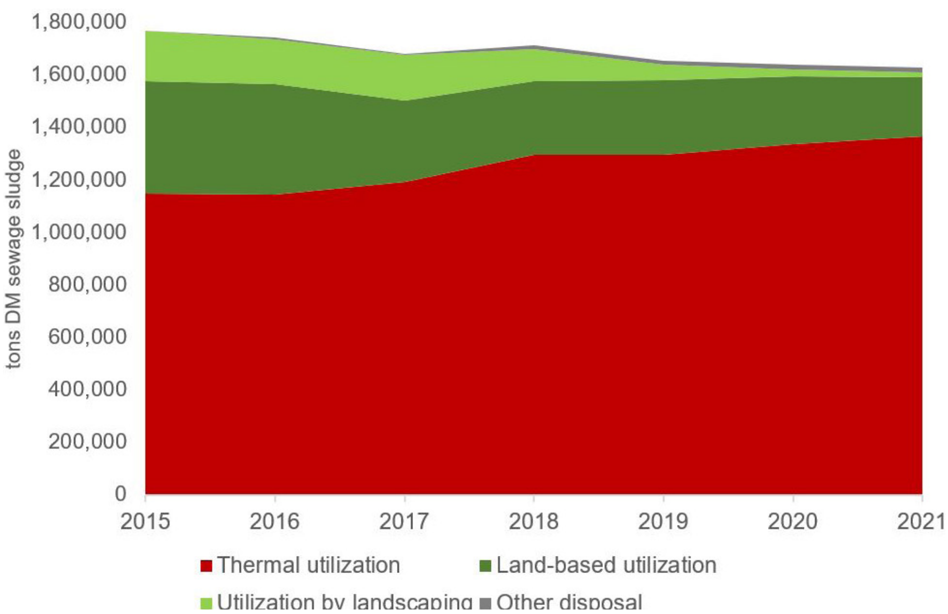

FIGURE 1: Development of the sewage sludge utilization in Germany 2015-2021 - data based on (DESTATIS, 2023).

TABLE 1: Composition of digested and dewatered sewage sludge according to Basse et al., 2012 and Roskosch et al., 2018.

Paramter	Unit	Range for sewage sludge	Limit values according to DüMV 2012 or AbfKlärV 2017 [mg/kg DM]
Water	Wt.-% (raw)	65 - 75	
Ash	Wt.-% (wf)	30 - 50	
LHV	MJ/Kg (raw)	1 - 2 / 10 - 12	
C	Wt.-% (wf)	33 - 50	
H	Wt.-% (wf)	3 - 4	
O	Wt.-% (wf)	10 - 20	
N	Wt.-% (wf)	2 - 6	
S	Wt.-% (wf)	0.5 - 1.5	
Fl	Wt.-% (raw)	aprox. 0.01	
Cl	Wt.-% (raw)	0.05 - 0.5	
P	g/kg (raw)	2 - 55	
Sb	mg/kg (raw)	5 - 30	
As	mg/kg (raw)	4 - 30	40
Pb	mg/kg (raw)	70 - 300	150
Cd	mg/kg (raw)	1.5 – 4.5	1.5
Cr	mg/kg (raw)	50 - 80	
Cu	mg/kg (raw)	300 - 350	
Mn	mg/kg (raw)	600 - 1,500	
Ni	mg/kg (raw)	30 - 35	80
Se	mg/kg (raw)	1 - 5	
Tl	mg/kg (raw)	0.2 - 0.5	1
V	mg/kg (raw)	10 - 100	
Hg	mg/kg (raw)	0.3 - 2.5	1
Zn	mg/kg (raw)	100 - 300	4,000
Sn	mg/kg (raw)	30 - 80	
AOX	mg/kg DM	200 - 400	400
PCB6	mg/kg DM	0.01 - 0.02	0.1
PCDD/dl-PCB	mg/kg DM	0.000005 – 0.0001	0.00003

2. HYDROTHERMAL CARBONIZATION OF SEWAGE SLUDGE

2.1 Operating conditions and reaction pathway

Hydrothermal carbonization (HTC) is a promising alternative for the management of sewage sludge and the process used moist waste as a reaction medium to convert biomasses into a lignite-like hydrochar. In contrast, other thermo-chemical conversion processes such as incineration and pyrolysis, are not suitable particularly for moist municipal and industrial organic waste like sewage sludge as it requires pre-drying (Wang et al., 2019B). A major advantage of the HTC technology is the possibility of processing the sewage sludge directly without further drying. As a result, the mechanically dewatered sewage sludge generated at WWTP could be processed directly (Wang et al., 2020). HTC has gained increasing interest from the research community and there are many scientific studies dealing with the hydrothermal treatment of sewage sludge like digested sludge (Huzeo et al., 2021), and raw sludge (Blach and Engelhart, 2020) or primary sludge (Danso-Boateng et al., 2015).

HTC converts sewage sludge into a carbonaceous product (hydrochar) while simultaneously recovering phosphorus from it by utilizing moisture present in sewage sludge as a catalyst, reactant, and solvent (Kruse et al., 2013). The sewage sludge is converted to a carbonaceous product under temperatures (180°C-280°C) and pressures below the saturation vapor pressure in several parallel or serial chemical reaction mechanisms (Reza et al., 2014a). During the process a series of reactions results in numerous dissolved intermediates. Polymers like carbohydrates first break down into simpler components (e.g., di- and monosaccharides) by hydrolysis. The hydrolysis of cellulose produces glucose and fructose. Lignin, on the other hand, is converted to phenol (Kruse et al., 2013). As the HTC process continues, various platform chemicals such as 5-HMF (5-(Hydroxymethyl)-2-furaldehyde), furfural, or levulinic acid are formed. Furthermore, Maillard reactions take place at certain reaction temperatures and reaction times, which contributes to the formation of hydrochar but also leads to high contents of total organic carbon (TOC) and total nitrogen (TN) in the resulting HTC process water (Xu et al., 2022; Djandja et al., 2021). The decisive reaction that leads to the formation of hydrochar is aldol condensation (Shi et al., 2019). The resulting hydrochar from HTC of sewage sludge is essentially hygienic, easy to dewater, free of pharmaceuticals, and expected to exhibit moderate energy density (Crocker et al., 2011; Saetea and Tippayawong, 2013). The yield of hydrochar produced depends on various reaction parameters, of which temperature and reaction time play a decisive role (Huezo et al., 2021; Djandja et al., 2021; Blach and Engelhart, 2020).

2.2 Fate and behavior of heavy metals and organic pollutants during HTC

As described in the introduction section, sewage sludge contains heavy metals along with organic pollutants. They are omnipresent in the environment and end up in sewage sludge from households and industries, among others (Roskosch et al., 2018). The sludge absorbs the heavy metals by passive sorption and active uptake through the accumulation of living cells (Geng et al., 2020).

Hydrothermal carbonization of sewage sludge has effects on the distribution of heavy metals. Wang et al., 2019A concluded that the heavy metal distribution depends on the reaction severity and suspected immobilization into the hydrochar. Wang et al, 2020 confirmed the assumption of redistribution of heavy metals in the liquid and solid phases. In particular, the metals Al, Fe, Zn, and Mn are immobilized in the char during the process under neutral conditions. Less than 5% of the heavy metals were detected in the resulting process water. However, acidic conditions result in a weaker enrichment of heavy metals into the solid phase. The immobilization depends mainly on the reaction temperature and reaction time (Wang et al., 2020). Furthermore, due to the immobilization of the heavy metals, the ecotoxicity is low (Liu et al., 2018; Wang et al., 2020). In addition to heavy metals, the fate of organic contaminants in sewage sludge, such as drug residues, is important. Vom Eyser et al., 2015, studied the removal of twelve pharmaceuticals (including ibuprofen and erythromycin) in sewage sludge during HTC. Seven of the pharmaceutical substances were either or the proportion in the hydrochar was reduced (39% to ≥ 97%) (vom Eyser et al., 2015). The heavy metals and organic impurities in the hydrochar from sewage sludge turn its suitability as fertilizer into a critical issue. However, vom Eyser et al., 2015 only investigated the fate of the pharmaceuticals in the respective hydrochar and not in the process water (PW). The PW can contain organic pollutants like phenols, aromatic compounds, alkenes, and pyrazines (Usman et al., 2019).

2.3 Process water as a by-product

The solid-liquid separation after hydrothermal carbonization of the sewage sludge produces two phases. On one side is the hydrochar and on the other side is the process water (PW). The PW is inorganically and organically polluted and has to be treated. Quicker and Weber, 2016 give ranges of values for COD from 4,900.00 - 78,000.00 mg l^{-1}, for BOD_5 1,700.00 - 42,000.00 mg l^{-1} and TOC 4,000.00 - 31,700.00 mg l^{-1}. If left untreated, it would be a burden on water bodies and thus on the environment (Leng et al., 2021). Particularly, the formation of persistent and toxic organic compounds during HTC that can leach into the PW remains a challenge. These substances are occasionally difficult to degrade biologically and may complicate the PW treatment. The composition of the PW depends primarily on the waste biomass used at the beginning and the reaction parameters (Woriescheck, 2019; Usman et al., 2019). Chen et al., 2019 found that PW from HTC of dewatered sewage sludge contains mainly proteins and short-chain fatty acids, but also poorly degradable compounds such as humic acids, melanoidins, nitrogen heterocycles, and phenols. A major challenge of PW management besides the actual contamination of the PW is the large quantities that would be produced during the industrial, continuous process of an HTC plant. Additionally, the legal requirements for PW treatment can also pose additional challenges. HTC-PW in Germany is categorized as wastewater and has to be treated and disposed of as such (§ 56 German Federal Water Act, WHG). This is done either to a direct discharge quality which allows direct discharge into the environment (§ 57 WHG) or to an indirect discharge quality which allows discharge and subsequent treatment in a municipal WWTP (§ 58 WHG). Respective requirements and limit values must be met for both alternatives.

The minimum requirement values for the discharge of wastewater into a water body or municipal WWTP are described in the German Waste Water Ordinance (AbwV) and the associated annexes. The treatment must be carried out according to the state of the art. For process waters from hydrothermal treatment, the requirements of Annex 27 "Treatment of waste by chemical and physical processes (CP plants) and waste oil regeneration" apply (AbwV 1997).

There is currently no continuous industrial implementation of HTC technology that includes process water management and treatment. One opportunity to manage the large quantities of PW is to reuse it as a partial flow in the HTC process and therefore save fresh water (if needed) and reduce the costs associated with it (Kambo et al., 2018). This step should be chosen to reduce costs and to benefit from the positive effects of recirculation. The aforementioned inorganic and organic load necessitate the use of PW treatment technologies. Biological processes such as anaerobic digestion proved to be feasible from various publications. Anaerobic degradation enables a reduction of chemical oxygen demand (COD) by up to 75% (Wirth et al., 2015). Another utilization pathway for PW is to recover nutrients from it. Malhotra and Garg, 2020 performed recovery of humic acids from PW using ammonium sulfate and were able to recover ~70% of humic acids. Subsequent growth trials showed positive effects (Malhotra and Garg, 2020). However, subsequent treatment of PW is necessary. Various providers of HTC solutions have their treatment technologies, like nanofiltration and reverse osmosis (Kläusli, 2014; Fettig et al., 2018) or aerobic treatment in a constructed wetland (Fettig et al., 2018). However, no data was available on the efficiency of these technologies and continuous large-scale operation is lacking. For a commercial realization of HTC technology, efficient and cost-effective processes for process water treatment are required.

2.4 Industrial readiness and industrial application

In addition to the mature technology, sufficient quantities of sewage sludge are required for the successful implementation of HTC technology for sewage sludge management and valorization in rural areas. The HTC technology must also be competitive with other thermal processes like incineration or pyrolysis to make its implementation successful. Reißmann et al., 2020 created a base model for HTC as a sewage sludge utilization method in Germany for 2030 assuming the currently best available techniques (BAT). In this model, 65,000 t FM sewage sludge per year is utilized (14,300 tons of dry matter per year with 22% DM). Due to the continuous industrial operation and the knowledge gained from it, learning effects can be observed in operational management. Nutrient recycling (e.g., phosphorus) is integrated in addition to the basic model. The authors concluded that production costs, phosphorus recovery, and process water treatment have a significant impact on the competitiveness of HTC compared to conventional processes such as incineration. According to this study, competitive production costs for the final HTC product must be less than € 325 per ton of hydrochar produced. Reißmann et al., 2020 point out that only the delivery of the product up to mono-incineration and not beyond was taken into account in this cost calculation. The HTC technology however has not yet (2023) been able to establish itself as a valorization process for the treated sewage sludge in Germany and especially for rural areas. The technology was temporarily tested on a sewage treatment plant in Düsseldorf but was discontinued after a test phase (Schnell et al., 2020). There are several methods for industrializing the technology at the moment, including multi-batch processes (quasi-continuous) and processes in tubular reactors (Blöhse, 2017; Quicker and Weber, 2016). There are some providers of commercial HTC solutions which includes TerraNova Energy, HTCycle, and SunCoal Industries (Aragón-Briceño et al., 2021).

3. HTC AND NUTRIENT RECOVERY

3.1 Need for phosphorus recovery and recovery technics

Phosphorus is a critical resource and a non-renewable essential element for all living organisms on the earth. The European Commission designated phosphorus as a "critical raw material" in 2014. (Wang et al., 2020; Aragón-Briceño et al., 2021). According to Krüger and Adam, 2015, more than 550,000 t/a of phosphorus was applied to German farmlands in 2015. The global phosphorus reserves are limited and the over-exploitation of natural reserves of phosphorus is anticipated to create severe problems in the next 50-100 years. Current phosphorus reserves are located in the US, Morocco, Jordan, China, and South Africa, assuming that Germany and Europe are entirely dependent on imports. The remaining phosphate reserves are however contaminated with heavy metals such as cadmium or uranium (Krüger and Adam, 2015). Various studies have assumed that the available phosphorus reserves will soon be exhausted (Pérez et al, 2021). Increasing population growth leads to higher consumption of energy and resources and to meet the phosphorus demand of the raising population, there is an immediate need for intelligent phosphorus recovery technologies and alternative sources of phosphorus. The integrated concerns such as; non-renewable nature, significance on global food security, and serious cost volatility, the future of phosphorus availability is drawing greater attention and poses an urgent need to find an alternative phosphorus source (Heckenmüller et al., 2014). As waste biomasses such as sewage sludge contain high phosphorus concentrations, several phosphorus recovery technologies are now available (Aragón-Briceño et al., 2021). The easiest way is land-based recycling with phosphorus being returned to the natural cycle. Rural areas in particular have benefited from land-based sewage sludge utilization. However, sewage sludge contains heavy metals and organic pollutants which makes the direct-recycling route more difficult (Aragón-Briceño et al., 2021). In this concern, the direct utilization of sewage sludge on agricultural land is prohibited. Several recovery technologies are based on the recovery of phosphorus from the sewage sludge ash, requiring incineration of the sewage sludge to produce phosphorus-rich ash (Kwapinski et al, 2021; Krüger and Adam, 2015). Hydrothermal carbonization in combination with subsequent phosphorus recovery would be an attractive option for rural regions without sewage sludge incineration (Kwapinski et al, 2021; Pérez et al, 2021).

3.2 Influence of acid utilization during HTC sewage sludge

3.2.1 Effects of Post- and Pre-Acid Treatment During HTC

HTC significantly reduces sewage sludge's acid buffering capacity due to the chemical reactions and thermochemical breakdown that occurs during the process. Carrying out acid leaching after HTC of sewage sludge, the majority of the H^+ in added acids will be utilized to induce the dissolution of Al-P and Fe-P and transformation of higher phosphorus from solid to liquid phase (Shettigondahalli et al., 2022). In contrast, when the acids are used during or before the HTC process, a significantly lower concentration of phosphorus will be transformed from the solid to the liquid phase. When acids are used before or during HTC, the majority of H^+ ions added in the form of acids are used to overcome buffering resistance provided by carbonates, cation exchange reactions, and organic matter. Due to the buffering agents present in the system, the available H^+ is greatly depleted before it can react with metal ions that hold phosphorus.

The presence of phosphate-precipitating metals such as Fe, Al, and Ca in sewage sludge plays a more significant role in determining the degree of conversion of phosphorus from solid to liquid (Huang et al., 2018; Ekpo et al., 2016). The presence of multivalent metal ions such as Al^{3+}, Ca^{2+}, Fe^{3+}, and Mg^{2+} during the HTC of sewage sludge contributes to the formation of phosphate with low solubility, which then facilitates the accumulation of phosphorus onto hydrochar. Similarly, the acid buffering resistance of the sewage sludge also regulates the phosphorus transformation from the solid to liquid phase upon the addition of acids. Sewage sludge suspension in general consists of a liquid and a solid phase. The solid and liquid phase of the sewage sludge contains several compounds with a high tendency to inhibit acidification (Shettigondahalli et al., 2022). The acid-inhibiting substance present in sewage sludge suspension tends to have different neutralizing points. Upon adding acids to sewage sludge suspension, H^+ would initially react with carbonates existing in the sewage sludge with relatively higher neutral points in the suspension. The additional H^+ will undergo a cation exchange after the carbonate in sewage sludge is exhausted. Later the presence of additional H^+ would be consumed in offsetting the buffering resistance provided by organic matter present in sewage sludge. In the end, the presence of any further H^+ would be utilized in the dissolution of aluminum phosphates (Al-P) and ferric phosphates (Fe-P) (Zhang et al., 2010; Bozkurt et al., 2000; Petzet et al., 2012). The HTC process however contributes considerably to reducing the acid buffering capacity of sewage sludge. By acid leaching the sewage sludge after HTC, there will be maximum utilization of added acids to induce the dissolution of Al-P and Fe-P and significantly increase the phosphorus-leaching from the solid to the liquid phase.

3.2.2 Recovery of Phosphorus from HTC-treated sewage sludge hydrochar

The utilization of H_2SO_4 and HCl at higher concentrations increases the mobilization of phosphorus (80-100%) from the solid to the liquid phase in hydrochar derived from sewage sludge (Pérez et al., 2021). Phosphorus leaching and the subsequent mobilization of heavy metals from hydrochars were investigated by Pérez et al., 2022, using oxalate and citrate across a pH range of 0 to 4. Oxalate resulted in nearly full P release from hydrochars at pH 1, whereas both oxalate and citric acids achieved P extraction efficiencies that were better than 75% even at the lowest pH. Acid leaching of the sewage sludge hydrochar results in a high phosphorus and multivalent cation recovery. Nevertheless, heavy metal concentrations must be monitored throughout the process since they affect the product's potential use as fertilizer. According to Gerner et al., hydrochar has an affinity towards heavy metals and will retain a significant portion of them, releasing very little into the liquid phase. The presence of a higher concentration of heavy metal in hydrochar makes the land-based application restricted under the German Fertilizer Ordinance. The most practical use option is then the energy recovery from hydrochar. HTC process also transfers around 50% of nitrogen from the solid to liquid phases, increasing hydrochar combustion performance in terms of NO_x emissions reduction (Gerner et al., 2021).

Calcium phosphate salt is a desirable product for recovering phosphorus from leachate. Calcium phosphate is similar to phosphate rock, the primary basic material for the fertilizer industry. The phosphorus product extracted from the effluent can be incorporated directly into the process as a by-product. The filtrate after leaching is believed to have a high ionic strength, which influences ion-association and solubility. Due to the decreased concentration of free ions caused by ion association, supersaturation may be required for the formation of solid deposits (Ehrnström, 2016). In order to recover all dissolved phosphorus, excess calcium ions must be introduced to the leachate. Increasing the pH could have a comparable effect.

4. CONCLUSIONS

The use of agriculture and landscaping as traditional treatment methods will soon be phased out in Germany due to new regulations on the management of sewage sludge (2029). Rural areas where sewage sludge is usually disposed of by agriculture and landscaping will be particularly challenged by the forthcoming situation. Hydrothermal carbonization (HTC) is receiving increased attention as an eco-friendly and promising technology for treating and managing sewage sludge without the need for drying. It is possible to effectively manage sewage sludge by using HTC technology not only by recovering phosphate but also by complying with German Sewage Sludge Ordinance requirements. HTC offers major potential for the valorization of sewage sludge and could be an interesting alternative for sewage sludge treatment and nutrient recovery in rural areas of Germany. Especially for nutrient recovery, HTC could be an opportunity to close loops. However, sufficient quantities of sewage sludge are needed to enable continuous operation. The HTC plant with subsequent process water treatment and nutrient recovery facilities should be built on the site of a WWTP to enable indirect discharge of pre-treated PW. The HTC process water is another challenge, as significant amounts are produced during continuous operation. The PW is organically contaminated and has to be treated before discharge into the environment. A functional and eco-economic concept for PW-management is necessary for a commercial implementation of HTC technology. Future research should focus on methods to minimize the amount of heavy metal in hydrochar and its use as fertilizer in accordance with the legal requirements of the German Fertilizer Ordinance and other relevant regulations. The technology has not yet (2023) been able to establish itself in an industrial continuously operated HTC plant for sewage sludge utilization in Germany. The technology readiness to manage and handle process water produced from HTC of sewage sludge is the major concern. A practical, dependable, and affordable process water disposal method is essential for the successful commercialization of HTC technology. Given the current rate of scientific development in the area, this is probably anticipated to happen in the near future.

AUTHORS CONTRIBUTION

Vicky Shettigondahalli Ekanthalu and Tommy Ender contributed equally to every aspect of this article until it was accepted and published online. Michael Nelles was responsible for reviewing and editing aspects of this article.

AKNOWLEDGEMENTS

The authors would like to thank Vidhi Singh for her kind assistance during the preparation of this article.

REFERENCES

Aragón-Briceño, C. I., Pozarlik, A. K., Bramer, E. A., Niedzwiecki, L., Pawlak-Kruczek H., Brem, G., 2021. Hydrothermal carbonization of wet biomass from nitrogen and phosphorus approach: A review, Renewable Energy, Volume 171, 401-415. https://doi.org/10.1016/j.renene.2021.02.109

Asghari, F. S., Yoshida, H., 2006. Acid-Catalyzed Production of 5-Hydroxymethyl Furfural from d-Fructose in Subcritical Water. Ind. Eng. Chem. Res. 2006, 45, 7, 2163–2173. https://doi.org/10.1021/ie051088y

AVA-CO2 Schweiz AG. 2014. "AVA-CO2 Achieves a Breakthrough in Phosphorus Recovery and Introduces the "AVA cleanphos" Process." September. Accessed May 2023. https://www.businesswire.com/news/home/20140910006209/en/AVA-CO2-Achieves-a-Breakthrough-in-Phosphorus-Recovery-and-Introduces-the-AVA-cleanphos-Process.

Basse, S., Buck, P., Domschke, T., Elstermann, N., Esser, R., Hanßen, H., Haselwimmer, T., Hiller, G., Jasper, M., Kappa, S., Kristkeitz, R., Lehrmann, F., Ludwig, P., Maurer, M., Ostertag, M., Peters, U., Pietsch, B., Steier, K., Werther, J., Wessel, M., Nath, C., Reifenstuhl, R., 2012. Merkblatt DWA-M 387 Thermische Behandlung von Klärschlämmen – Mitverbrennung in Kraftwerken. DWA Deutsche Vereinigung für Wasserwirtschaft, Abwasser und Abfall e. V.

Blach, T., Engelhart, M., 2020. Optimizing the Hydrothermal Carbonization of Sewage Sludge—Response Surface Methodology and the Effect of Volatile Solids. Water, 13 (9), p. 1225. https://doi.org/10.3390/w13091225

Blöhse, D., 2017. Hydrothermale Karbonisierung – Nutzen dieser Konversionstechnik für die optimierte Entsorgung feuchter Massenreststoffe (Doctoral dissertation). University Essen, Germany: Duisburg-Essen. Retrieved from https://duepublico2.uni-due.de/receive/duepublico

Bozkurt, S., Moreno, L., Neretnieks, I., 2000. Long-Term Processes in Waste Deposits. Science of the Total Environment, 250, 101-121. https://doi.org/10.1016/S0048-9697(00)00370-3

Buttmann, M., 2023. TerraNova®ultra project for sewage sludge and biowaste in Poland. Retrieved from https://www.terranova-energy.com/en/terranovaultra-project-for-sewage-sludge-and-biowaste-in-poland/ Last retrieved 31.05.2023

Chen, H., Rao, Y., Cao, L., Shi, Y., Hao, S., Lou, G., Zhang, S., 2019. Hydrothermal conversion of sewage sludge: Focusing on the characterization of liquid products and their methane yields. Chemical Engineering Journal, Volume 357, 2019, Pages 367-375. https://doi.org/10.1016/j.cej.2018.09.180

Crocker, M., 2011. Thermo chemical Conversion of Biomass to Liquid Fuels and chemicals. Royal Society of Chemistry Publishing. https://doi.org/10.1039/9781849732260
Danso-Boateng, E., Sharma, G., Wheatly, A. D., Martin, S. J., Holdich, R. G., 2015. Hydrothermal carbonisation of sewage sludge: Effect of process conditions on product characteristics and methane production. Bioresource Technology, Volume 177, 2015. 318-327. https://doi.org/10.1016/j.biortech.2014.11.096
Djandja, O. S., Yin, L., Wang, Z.-C., Duan, P.-G., 2021. From wastewater treatment to resources recovery through hydrothermal treatments of municipal sewage sludge: A critical review. Process Safety and Environmental Protection, 151, 101–127. https://doi.org/10.1016/j.psep.2021.05.006
Ehrnström, Matilda Sirén. 2016. "Recovery of Phosphorus from HTC Converted Municipal Sewage Sludge." MASTER OF SCIENCE THESIS, Department of Civil, Environmental and Natural Resources Engineering, Luleå University of Technology.
Ekpo, U., Ross, A. B., Camargo-Valero, M. A., Fletcher, L. A., 2016. Influence of pH on hydrothermal treatment of swine manure: Impact on extraction of nitrogen and phosphorus in process water. Bioresource Technology, Volume 214, 2016, Pages 637-644. https://doi.org/10.1016/j.biortech.2016.05.012
Geng, H., Xu, Y., Zheng, L., Gong, H., Dai, L., Dai, X., 2020. An overview of removing heavy metals from sewage sludge: Achievements and perspectives. Environmental Pollution, Volume 266, Part 2, 115375. https://doi.org/10.1016/j.envpol.2020.115375
Gerner, G., Meyer, L., Wanner, R., Keller, T., Krebs, R. (2021): Sewage Sludge Treatment by Hydrothermal Carbonization: Feasibility Study for Sustainable Nutrient Recovery and Fuel Production. Energies 2021, 14, 2697. https://doi.org/10.3390/en14092697
Heckenmüller, M., Narita, D., Klepper, G., 2014. Global availability of phosphorus and its implications for global food supply: An economic overview, Kiel Institute for the World Economy (IfW), Kiel.
Huang, R, Fang, C., Zhang, B., Tang, Y., 2018. Transformations of Phosphorus Speciation during (Hydro)thermal Treatments of Animal Manures. Environ. Sci. Technol. 52 (5): 3016-3026. https://doi.org/10.1021/acs.est.7b05203
Huezo, L., Vasco-Correa, J., Shah, A., 2021. Hydrothermal carbonization of anaerobically digested sewage sludge for hydrochar production. Bioresource Technology Reports, Volume 15, 2021, 100795. https://doi.org/10.1016/j.biteb.2021.100795
Kambo, H. S., Minaret, J., Dutta, A. (2018): Process Water from the Hydrothermal Carbonization of Biomass: A Waste or a Valuable Product?. Waste Biomass Valor 9, 1181–1189 (2018). https://doi.org/10.1007/s12649-017-9914-0
Klärschlammverordnung (AbfKlärV) vom 27. September 2017 (BGBl. I S. 3465), die zuletzt durch Artikel 137 der Verordnung vom 19. Juni 2020 (BGBl. I S. 1328) geändert worden ist
Krüger, O., Adam, C., 2015. Recovery potential of German sewage sludge ash, Waste Management, Volume 45, 2015, 400-406. https://doi.org/10.1016/j.wasman.2015.01.025
Kruse, A., Funke, A., Titirici, M.-M., 2013. Hydrothermal conversion of biomass to fuels and energetic materials. Current Opinion in Chemical Biology, Volume 17, Issue 3, 2013, 515-521. https://doi.org/10.1016/j.cbpa.2013.05.004
Kwapinski, W., Kolinovic, I., Leahy, J.J., 2021. Sewage Sludge Thermal Treatment Technologies with a Focus on Phosphorus Recovery: A Review. Waste Biomass Valor 12, 5837–5852. https://doi.org/10.1007/s12649-020-01280-2
Langenohl, T., 2015. Auswirkungen der sich verändernden Rahmenbedingungen auf die Entsorgungssicherheit für Klärschlamm. KA Korrespondenz Abwasser, Abfall, Jahrgang 62, Heft 3, 249-256. http://dx.doi.org/10.3242/kae2015.03.003
Leng, L., Zhang, W., Leng, S., Chen, J., Yang, L., Li, H., Jiang, S., Huang, H., 2020: Bi.oenergy recovery from wastewater produced by hydrothermal processing biomass_Progress, challenges, and opportunities. Science of The Total Environment, Volume 748, 2020, 142383, https://doi.org/10.1016/j.scitotenv.2020.142383
Li, F., Ji, W., Chen, Y., Gui, X., Li, J., Zhao, J., Zhou, C., 2021. Effect of Temperature on the Properties of Liquid Product from Hydrothermal Carbonization of Animal Manure and Function as a Heavy Metal Leaching Agent in Soil. Water Air Soil Pollut 232, 189 (2021). https://doi.org/10.1007/s11270-021-05134-y
Libra, J. A., Ro, K. S., Kammann, C., Funke, A., Berge, N. D., Neubauer, Y., Titirici, M.-M., Fühner, C., Bens, O., Kern, J., 2011. Hydrothermal carbonization of biomass residuals: a comparative review of the chemistry, processes and applications of wet and dry pyrolysis. Biofuels 2(1):71–106. https://doi.org/10.4155/bfs.10.81
Liu, T., Liu, Z., Zheng, Q., Lang, Q., Xia, Y., Peng, N., Gai, C., 2018. Effect of hydrothermal carbonization on migration and environmental risk of heavy metals in sewage sludge during pyrolysis. Bioresour Technol 247:282–290. https://doi.org/10.1016/j.biortech.2017.09.090
Malhotra, M. and Garg, A., 2020. Hydrothermal carbonization of centrifuged sewage sludge: Determination of resource recovery from liquid fraction and thermal behaviour of hydrochar. Waste Management, Volume 117, November 2020, 114-123. https://doi.org/10.1016/j.wasman.2020.07.026
Mix-Spagl, K., 2017. Die neue Klärschlammverordnung – Was müssen Betreiber beachten? Wwt-Online.de - Special Klärschlamm - Recht & Gesetz, 2017, 10, 15-17.
Montag, D., Adam, C., Baumann, P., Frank, D., Kabbe, C., Klein, D., Meyer, C., Mocker, M., Morf, L., Pinnekamp, J., Roskosch, A., Schaum, C., Schneichel, W., Schneider, Y., Wett, M., Arnold, U., Heidecke, P., Sichler, T., 2022. Rechtliche Vorgaben der Klärschlammverordnung und deren Auswirkungen auf die Phosphor-Rückgewinnung. KA Korrespondenz Abwasser, Abfall, Jahrgang 69, Heft 5, 406-414. http://dx.doi.org/10.3242/kae2022.05.005
Pérez, C., Boily, J. F., Jansson, S., 2021. Acid-Induced Phosphorus Release from Hydrothermally Carbonized Sewage Sludge. Waste Biomass Valor 12, 6555–6568. https://doi.org/10.1007/s12649-021-01463-5
Pérez, Carla, Jean-François Boily, Nils Skoglund, Stina Jansson, and Jerker Fick. 2022. "Phosphorus release from hydrothermally carbonized digested sewage sludge using organic acids." Waste Management 60-69. https://doi.org/10.1016/j.wasman.2022.07.023
Petzet, S., Peplinski, B., Cornel, P., 2012. On wet chemical phosphorus recovery from sewage sludge ash by acidic or alkaline leaching and an optimized combination of both. Water Res 46: 3769-3780. https://doi.org/10.1016/j.watres.2012.03.068
Quicker, P. and Weber, K., 2016. Biokohle - Herstellung, Eigenschaften und Verwendung von Biomassekarbonisaten. Springer-Verlag, Berlin
Reißmann, D., Thrän, D., Blöhse, D., Bezama, A., 2020. Hydrothermal carbonization for sludge disposal in Germany: A comparative assessment for industrial-scale scenarios in 2030. Journal of Industrial Ecology, 25: 720– 734. https://doi.org/10.1111/jiec.13073
Reza, M. T., Andert, J., Wirth, B., Busch, D., Pielert, J., Lynam, J., Mumme, J., 2014. Review Article: Hydrothermal Carbonization of Biomass for Energy and Crop Production. Applied Bioenergy 2014. 1:11-29. http://dx.doi.org/10.2478/apbi-2014-0001
Reza, M. T., Uddin, M. H., Lynam, J. G., 2014. Hydrothermal carbonization of loblolly pine: reaction chemistry and water balance. Biomass Conv. Bioref. 4, 311–321. https://doi.org/10.1007/s13399-014-0115-9
Roskosch, A., Heidecke, Patric, Bannick, C., Brandt, S., Bernicke, M., Dienemann, C., Gast, M., Hofmeier, M., Kabbe, C., Schwirn, K., Vogel, I., Völker, D., Wiechmann, B., 2018. Klärschlammentsorgung in der Bundesrepublik Deutschland (sewage sludge disposal in Germany). Umweltbundesamt, Dessau-Roßlau.
Saetea, P., Tippayawong, N., 2013. Recovery of Value-Added Products from Hydrothermal Carbonization of Sewage Sludge. Chemical Engineering, Volume 2013. https://doi.org/10.1155/2013/268947
Schnell, M., Horst, T., Quicker, P., 2020. Thermal treatment of sewage sludge in Germany: A review. Journal of Environmental Management, Volume 263, 110367. https://doi.org/10.1016/j.jenvman.2020.110367
Shettigondahalli, E. V., Morscheck, G., Narra, S., Nelles, M., 2020. Hydrothermal Carbonization—A Sustainable Approach to Deal with the Challenges in Sewage Sludge Management. In: Ghosh, S. (eds) Urban Mining and Sustainable Waste Management. Springer, Singapore. https://doi.org/10.1007/978-981-15-0532-4_29
Shettigondahalli, E. V., Narra, S., Ender, T., Antwi, E., Nelles, M., 2022. Influence of Post- and Pre-Acid Treatment during Hydrothermal Carbonization of Sewage Sludge on P-Transformation and the Characteristics of Hydrochar. Processes; 10(1):151. https://doi.org/10.3390/pr10010151
Shi, N., Liu, Q., He, X., Wang, G., Chen, N., Peng, J., Longlong Ma, 2019. Molecular Structure and Formation Mechanism of Hydrochar from Hydrothermal Carbonization of Carbohydrates. Energy Fuels 2019, 33, 10, 9904–9915. https://doi.org/10.1021/acs.energyfuels.9b02174
Statistisches Bundesamat, DESTATIS, 2023. Klärschlammentsorgung nach Bundesländern. https://www.destatis.de/DE/Themen/Gesellschaft-Umwelt/Umwelt/Wasserwirtschaft/Tabellen/liste-klaerschlammverwertungsart.html

Usman, M., Chen, H., Chen, K., Ren, S., Clark, J. H., Fan, J., Luo, G., Zhang, S. (2019): Characterization and utilization of aqueous products from hydrothermal conversion of biomass for bio-oil and hydro-char production: a review. Green Chem., 2019,21, 1553-1572. https://doi.org/10.1039/C8GC03957G

vom Eyser, C., Palmu, K., Schmidt, T.C., Tuerk, J., 2015. Pharmaceutical load in sewage sludge and biochar produced by hydrothermal carbonization. Sci. Total Environ. 537, 180–186. https://doi.org/10.1016/j.scitotenv.2015.08.021

Wang, H., Yang, Z., Li, X., 2020. Distribution and transformation behaviors of heavy metals and phosphorus during hydrothermal carbonization of sewage sludge. Environ Sci Pollut Res 27, 17109–17122. https://doi.org/10.1007/s11356-020-08098-4

Wang, L., Chang, Y., Liu, Q., 2019A. Fate and distribution of nutrients and heavy metals during hydrothermal carbonization of sewage sludge with implication to land application, Journal of Cleaner Production, Volume 225, 2019, 972-983. https://doi.org/10.1016/j.jclepro.2019.03.347

Wang, L., Chang, Y., Li, A., 2019B. Hydrothermal carbonization for energy-efficient processing of sewage sludge: A review. Renewable and Sustainable Energy Reviews, Volume 108, 2019, 423-440. https://doi.org/10.1016/j.rser.2019.04.011

Wilson, C.-A., Novak, J.-T., 2009. Hydrolysis of macromolecular components of primary and secondary wastewater sludge by thermal hydrolytic pretreatment, Water Research, Volume 43, Issue 18, 2009, 4489-4498. https://doi.org/10.1016/j.watres.2009.07.022

Wirth, B., Reza, M. T., Mumme, J., 2015. Influence of digestion temperature and organic loading rate on the continuous anaerobic treatment of process liquor from hydrothermal carbonization of sewage sludge. Bioresource Technology, Volume 198, 2015, Pages 215-222. https://doi.org/10.1016/j.biortech.2015.09.022

Woriescheck, T., 2019. Charakterisierung, Aufreinigung und Wertstoffgewinnung von Prozesswasser der Hydrothermalen Carbonisierung. (Doctoral dissertation). University of Oldenburg, Germany: Oldenburg. Retrieved from https://plus.orbis-oldenburg.de/primo-explore/fulldisplay?docid=49GBVUOB_ALMA51338789630003501&context=L&vid=UB_V1&lang=de_DE&search_scope=UB_all&adaptor=Local%20Search%20Engine&tab=default_tab&query=any,contains,49GBVUOB_ALMA51338789630003501

Xu, Z.-X., Ma, X.-Q., Zhou, J., Duan, P.-G., Ahmad, A., Luque, R., 2022. The influence of key reactions during hydrothermal carbonization of sewage sludge on aqueous phase properties: A review. Journal of Analytical and Applied Pyrolysis, Volume 167, 2022, 105678. https://doi.org/10.1016/j.jaap.2022.105678

Zhang, H.-Y., Ju, Y.-Y., Fan, Z.-M., Wang, B., 2010. Acid buffer capacity of sewage sludge barrier for immobilization of heavy metals. Huan Jing Ke Xue. 2010 Dec;31(12):2956-64. Chinese. PMID: 21360886